(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 0 908 174 A2

(12) EUROPEAN PATENT APPLICATION

(43) Date of publication:
14.04.1999 Bulletin 1999/15

(51) Int Cl.6: A61K 7/46, G01N 30/00

(21) Application number: 98306584.8

(22) Date of filing: 18.08.1998

(84) Designated Contracting States:
AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU MC NL PT SE
Designated Extension States:
AL LT LV MK RO SI

(30) Priority: 18.09.1997 US 933599
30.10.1997 US 961561
23.04.1998 US 64732

(71) Applicant: INTERNATIONAL FLAVORS & FRAGRANCES INC.
New York New York 10019 (US)

(72) Inventors:
- Shefer, Adi
Middlesex, New Jersey 08816 (US)
- McDermott, Keith J.
Somerset, New Jersey 08805 (US)
- Shefer, Shmuel David
Middlesex, New Jersey 08816 (US)
- Tan, Chee-Teck
Monmouth, New Jersey 07748 (US)

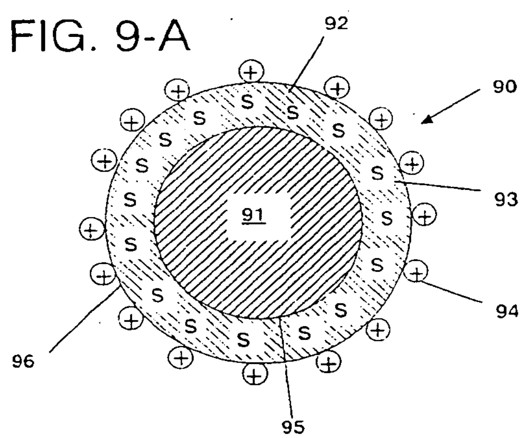
(74) Representative: Brown, John David et al
FORRESTER & BOEHMERT
Franz-Joseph-Strasse 38
80801 München (DE)

(54) Compositions for fragrancing at least one perfumable material and fragrance diffusion evaluation apparatus

(57) Described are controlled, time-release microparticulate active and bioactive compositions (including perfuming compositions) for targeted delivery to services such as skin, hair and fabric and the environment proximate thereto, where the active and bioactive materials have a calculated $\log_{10}P$ values of between 1 and 8 (**P** being the n-octanol-water partition coefficient). Such compositions include the active or bioactive material in single phase, solid solution in a wax or polymer matrix also having coated thereon and/or containing a compatible surfactant. Also described are processes and apparatus for preparing such compositions and processes for using same. Furthermore, certain component(s) of the aforementioned compositions in combination with one another are novel, and other components have novel uses in increasing fragrance substantivity.

Also described is fragrance diffusion evaluation apparatus for testing the diffusivity, odor character and odor intensity of a fragrance material and a process for using same. The apparatus includes at least one hollow container means having a test sample suspended therein, air flow means for passing air through the inside of the container means and an exit port for the air flow where the intensity and aroma character are measured as a function of time and as a function of temperature. The sample is weighed initially and at time intervals while the air is flowing through the cylinders at a fixed or variable rate.

FIG. 9-A



Printed by Jouve, 75001 PARIS (FR)

(Cont. next page)

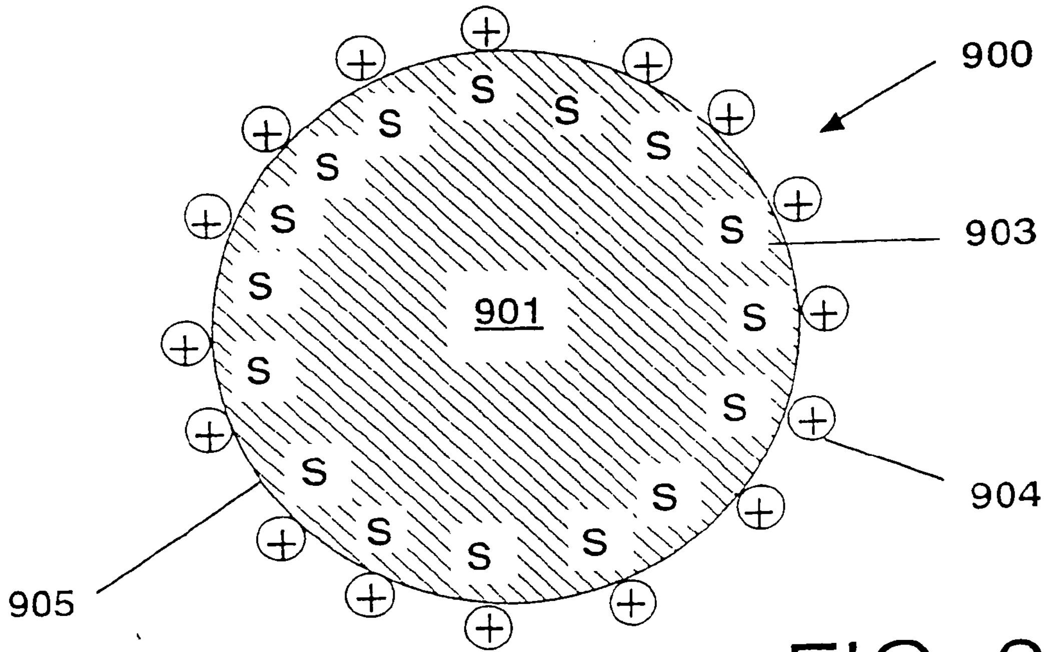
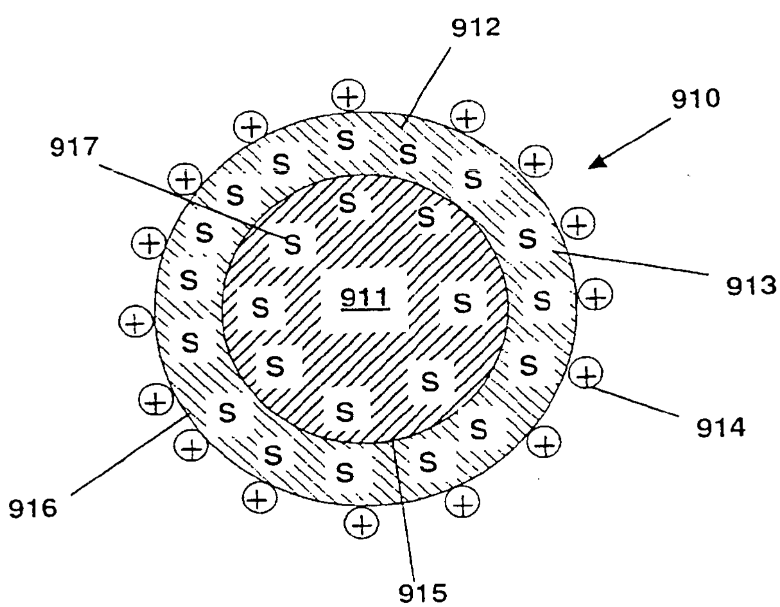


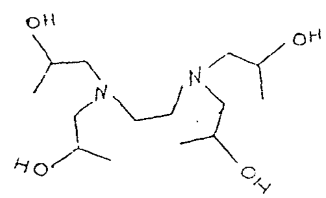
FIG. 9-B

FIG. 9-C



Description

[0001] Our invention concerns controlled time-release microparticulate active and bioactive compositions (including perfuming compositions) for targeted delivery to surfaces such as skin, hair and fabric and the environment proximate thereto, where the active and bioactive materials have a calculated $\log_{10}P$ of between 1 and 8 (**P** being the n-octanol-water partition coefficient). Such compositions include the active or bioactive material in single phase solid solution in a wax or polymer matrix also having coated thereon and/or containing a compatible surfactant. Our invention is also directed to processes and apparatus for preparing such compositions and processes for using same. Furthermore, certain component(s) of the above-mentioned compositions in combination with one another are novel; both combinations containing partially hydrolyzed polyvinyl acetate having a degree of hydrolysis of between about 73% up to about 99% and having a molecular weight in the range of from about 5,000 up to about 67,000. Our invention is also directed to novel compositions having high perfume substantivity including the compound: tetra(2-hydroxypropyl) ethylenediamine having the structure:



[0002] More particularly, our invention is directed to a composition for effecting the targeted delivery of a bioactive or active substance to a substantially solid surface comprising at least one substantially ellipsoidal hydrophilic particle having a continuous outer surface and an internal matrix volume consisting essentially of:

(i) a single phase solid solution of a matrix material which is in the alternative at least one of a hydrophobic polymer and/or at least one hydrophobic wax, each of which polymer and wax has a melting point in the range of from about 35°C up to about 120°C at 1 atmosphere pressure, having dissolved therein at least one active or bioactive substance (for example, a fragrance material) which is hydrophobic, said solid solution having an outer surface and an internal matrix volume; and

(ii) proximate to substantially to the entirety of said outer surface a substantially hydrophilic surfactant.

[0003] The active or bioactive material, such as a fragrance material, having a calculated $\log_{10}P$ in the range of from about 1 up to about 8 wherein P is the partition coefficient of the active or bioactive material between n-octanol and water; with the hdyrophobic particle having an outside diameter in the range of from about 0.05 up to about 20 microns; the concentration of active or bioactive material in the polymer or the wax being from about 5% up to about 60% by weight of the particle; the weight percent of the surfactant being from about 0.01% up to about 5% by weight of the particle; with the wax, the surfactant and the polymer each being nonreactive with the bioactive or active material and one another.

[0004] A preferred composition of our invention is one where the permeation rate of the active or bioactive material, such as the fragrance material, through the wax or the polymer is in the range of from about $10^{-8}\frac{mg \cdot mm}{cm^2 \cdot min}$ up to about $8\times10^{-3}\frac{mg \cdot mm}{cm^2 \cdot min}$ as determined by the IFF permeation test as more fully described herein in the "DETAILED DESCRIPTION OF THE DRAWINGS" section, infra.

[0005] As stated, supra, proximate to substantially the entirety of the outer surface of the substantially ellipsoidal hydrophobic particle is a substantially hydrophilic surfactant. More specifically, the following three cases exist concerning the location of the surfactant:

(a) the substantially hydrophilic surfactant may be substantially entirely coded on and fixedly bonded to the entirety of the outer surface of the single phase solid solution in the form of a continuous submicron layer of surfactant; or

(b) the substantially hydrophilic surfactant may be located proximate to and immediately, substantially beneath the entirety of the outer surface of the solid solution and substantially within the said internal matrix volume; and

(c) the substantially hydrophilic surfactant is both (a) substantially, entirely coated on and fixedly bonded to the

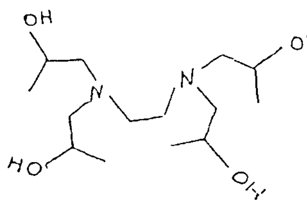
entirety of the outer surface of the single phase solid solution in the form of a continuous submicron layer of surfactant and (b) located proximate to and immediately, substantially beneath the entirety of the outer surface of the solid solution and substantially within the internal matrix volume

[0006] With respect to the surfactant, the surfactant may be a cationic surfactant, and the particle would therefore be positively charged, the surfactant may be an anionic surfactant, and the particle would be negatively charged; the surfactant could be a nonionic surfactant, and the particle would have a neutral charge, and the surfactant is a zwitterionic surfactant, and the particle has a variable charge.

[0007] Examples of surfactants particularly preferred in the practice of our invention are as follows

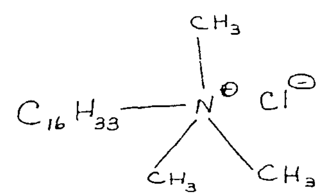
(a) the cationic modified starch, RediBOND® 5320 (trademark of the National Starch Company of Bridgewater, New Jersey), in admixture with partially hydrolyzed polyvinyl acetate having a degree of hydrolysis of between about 73% up to about 99% and having a molecular weight in the range of from about 5,000 up to about 67,000;

(b) the substance tetra(2-hydroxypropyl) ethylenediamine (marketed, for example, as QUADROL® Polyol, and having the structure.



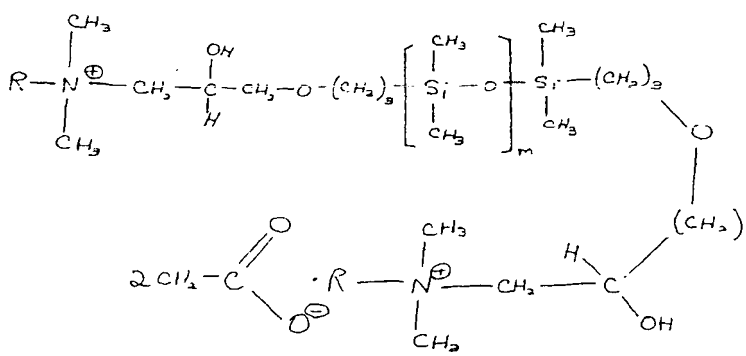
);

(c) cetyl trimethyl ammonium halide, including cetyl trimethyl ammonium chloride having the structure:



;

(d) a quaternary ammonium polysilane derivative having the structure:



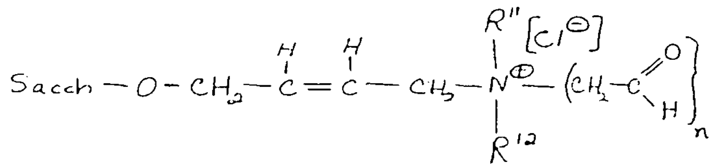
wherein **R** is the moiety having the structure:

$$CH_3\text{-}[CH_2]_x\text{-};$$

wherein **x** is an integer of from 10 up to 100 and **m** is an integer of from 10 up to 100 in admixture with partially

hydrolyzed polyvinyl acetate being hydrolyzed to the extent of from about 73% up to about 99% and having a molecular weight in the range of from about 5,000 up to about 67,000; and

(e) the cationic polysaccharide derivative defined according to the structure:



wherein **n** is an integer of from 1 up to 3; $\mathbf{R}^{11}$ and $\mathbf{R}^{12}$ are independently an alkyl, aryl, aralkyl or alkaryl group when **n** is 1; $\mathbf{R}^{11}$ or $\mathbf{R}^{12}$ is one of the groups when **n** is 2; or $\mathbf{R}^{11}$ and $\mathbf{R}^{12}$ are not present when **n** is 3; and wherein the moiety "SACCH" represents a starch or cellulose moiety.

[0008] The weight ratio of cationic modified starch:partially hydrolyzed polyvinyl acetate, is preferably in the range of from about 2:1 up to about 1:2, with a ratio of 1:2 being preferred. The weight ratio of the quaternary ammonium polysilane derivative:partially hydrolyzed polyvinyl acetate, is also preferably in the range of from about 2:1 up to about 1:2, with a weight ratio of 1:2 being preferred.

[0009] The mixtures of the cationic modified starch and partially hydrolyzed polyvinyl acetate as well as the quaternary ammonium polysilane derivative and partially hydrolyzed polyvinyl acetate are novel mixtures.

[0010] The matrix material which may be at least one hydrophobic polymer and/or at least one hydrophobic wax useful in the practice of our invention is preferably at least one of the following materials:

(a) polyamides having a molecular weight in the range of from about 6, 000 up to about 12, 000, for example, MACROMELT® 6030 manufactured by the Henkel Ag. of Dusseldorf, Germany (other examples being set forth in Lindauer, et al, U.S. Letters Patent No. 4,184,099 issued on January 15, 1980, the specification for which is incorporated by reference herein and including the VERSALON® line of polyamide polymers manufactured by the Henkel Corporation of Minneapolis, Minnesota) ;

(b) synthetic and natural carnauba wax;

(c) synthetic and natural candelilla wax;

(d) mixtures of cetyl palmitate (marketed, for example, as CUTINA® wax) with carnauba wax;

(e) mixtures of cetyl palmitate and candelilla wax;

(f) ozokerite wax;

(g) ceresin wax; and

(h) low density polyethylene wax having a molecular weight in the range of from about 500 up to about 6,000.

[0011] Different combinations of waxes and surfactants are preferred for different fragrance compositions having different overall calculated $\mathbf{log_{10}P}$ for different applications, for example, hair care or fabric care.

[0012] The maximum vapor pressure for the active or bioactive material in the composition of our invention should be 4.1 mm/Hg at 30°C. In the event that the active material is a fragrance material, it is preferred that when the fragrance material has topnote components, middle note components and bottom note components, the vapor pressure ranges for each of these three groups of components should be as follows:

(a) with respect to the topnote components, the vapor pressure range should be from 0.0001 mm/Hg up to 0.009 mm/Hg at 25°C;

(b) with respect to the middle note components, the vapor pressure range of the middle note components should be from 0.01 mm/Hg up to 0.09 mm/Hg at 25°C; and

(c) with respect to the bottom note components, the vapor pressure range of the bottom note components should be from 0 1 mm/Hg up to 2 0 mm/Hg at 25°C

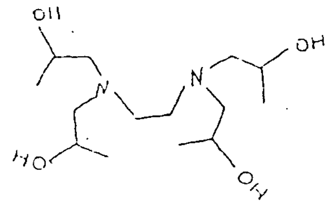
[0013] An example of such a fragrance as described, supra, is as follows:

Type of Note	Component	Vapor Pressure mm/Hg at 25°C
bottom note	TONALID® (trademark of Givaudan SA of Geneva, Switzerland)	0.0001
bottom note	hexyl cinnamic aldehyde	0.0003
bottom note	cis-3-hexenyl salicylate	0 0008
bottom note	ISO E SUPER® (trademark of International Flavors & Fragrances Inc of New York, NY)	0.002
bottom note	peach aldehyde coeur	0.002
bottom note	LILIAL® (trademark of Givaudan, Inc. of Clifton, NJ)	0 003
bottom note	cyclamal	0.004
bottom note	β-ionone	0 006
bottom note	γ-methyl ionone	0.006
bottom note	citronellol	0.009
bottom note	methyl nonyl acetaldehyde	0.009
middle note	allyl cyclohexyl propane	0.01
middle note	α-terpineol	0.02
middle note	1-borneol	0.02
middle note	dipropylene glycol	0.02
middle note	hyacinth extract	0.02
middle note	β-phenyl ethyl alcohol	0.02
middle note	VERTENEX® HC (trademark of International Flavors & Fragrances Inc. of New York, NY)	0.03
middle note	linalool	0.05
middle note	allyl amyl glycolate	0.07
middle note	linalyl acetate	0 07
middle note	dihydromyrcenol	0.09
middle note	isobornyl acetate	0.09
middle note	methyl chavicol	0.09
top note	benzyl acetate	0.1
top note	camphor	0.1
top note	styralyl acetate	0.1
top note	ALDEHYDE AA Triplal™ (trademark of International Flavors & Fragrances Inc. of New York, NY)	0.3
top note	eucalyptus oil	1.7
top note	cis-3-hexenyl acetate	2.0

[0014] The particles of the composition of our invention may contain or have coated thereon (or both) surfactants having (i) a sufficient charge per molecule of surfactant and (ii) a sufficient concentration of surfactant in each particle so that the electrostatic charge density on the surface of each particle will be sufficient to cause adherence of the

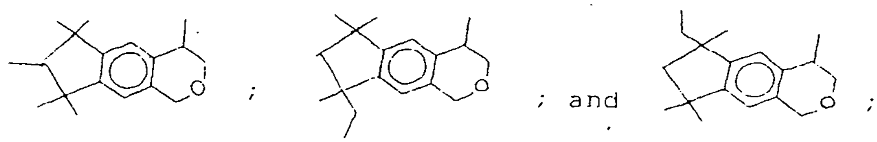
particle to a given surface, such as hair, mammalian skin or a fabric.

[0015] While using the material, tetra(2-hydroxypropyl) ethylenediamine having the structure:

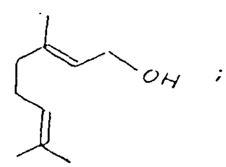


as a surfactant , we have determined that this material is also surprisingly useful in increasing substantivity of fragrances and aroma chemicals when the rate ratio of tetra (2-hydroxypropyl) ethylenediamine:fragrance material is from about 2:15 up to about 4:5. Examples of materials for which the fragrance substantivity will be increased to an extent of greater than about 50% are as follows:

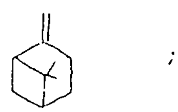
(a) GALAXOLIDE®, a mixture of compounds having the structures:



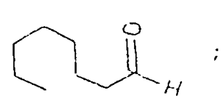
(b) geraniol having the structure:



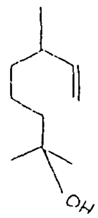
(c) β-pinene having the structure:



(d) n-octanal having the structure:

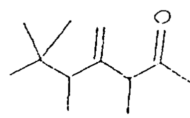


(e) dihydromyrcenol having the structure:



;

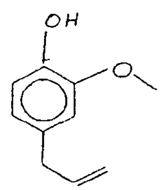
(f) KOAVONE® (trademark of International Flavors & Fragrances Inc of New York, NY) having the structure:



;

and

(g) eugenol having the structure.

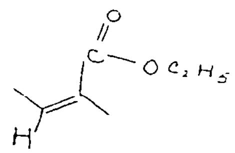


.

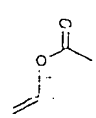
[0016] As indicated, supra, the range of permeation rates of the active and bioactive materials through the wax or polymer of the solid solution-containing particles of our invention is in the range of from about $10^{-8}\frac{mg \cdot mm}{cm^2 \cdot min}$ up to about $8\times10^{-3}\frac{mg \cdot mm}{cm^2 \cdot min}$. Specifically, the following materials having the following calculated **$\log_{10}P$** also have the following permeation rates through various waxes and polymers useful in the practice of our invention:

Aroma Chemical	Matrix Material	Permeation Rate $\frac{mg - mm\ thickness}{cm^2\ area - minute}$	Calculated $\log_{10}P$
β-pinene	carnauba wax	2.8×10^{-4}	4.6
β-pinene	polyethylene wax having a molecular weight of 500	8.2×10^{-4}	4.6
β-pinene	polyamide (MACROMELT® 6030)	2.1×10^{-4}	4.6

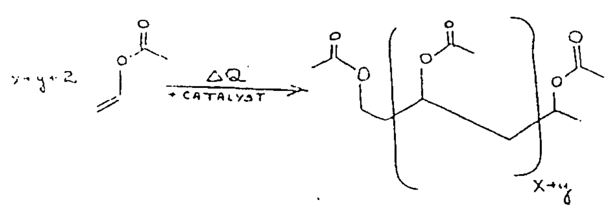
continued-

Aroma Chemical	Matrix Material	Permeation Rate $\frac{mg - mm\ thickness}{cm^2\ area - minute}$	Calculated $\log_{10}P$
ethyl tiglate having a structure: 	carnauba wax	4.4×10^{-4}	2.0
ethyl tiglate	polyethylene wax having a molecular weight of 500	2.0×10^{-5}	2.0
ethyl tiglate	polyamide (MACROMELT® 6030)	4.2×10^{-4}	2.0
Fragrance mixture S-1	carnauba wax	5.3×10^{-4}	--
Fragrance S-1	candelilla wax	2.7×10^{-4}	--
Fragrance S-1	polyamide (MACROMELT® 6030)	3.8×10^{-3}	--
Fragrance S-1	polyethylene wax (molecular weight 500)	6.3×10^{-4}	--
Fragrance O-1	carnauba wax	1.4×10^{-3}	--
Fragrance O-1	candelilla wax	7×10^{-4}	--

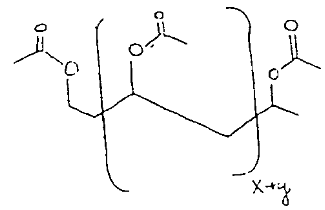
[0017] In practicing our invention, the partially hydrolyzed polyvinyl acetate, also termed "polyvinyl alcohol" where the polyvinyl acetate is hydrolyzed to an extent of from about 73% up to about 99%, is prepared by means of any of Examples I-XIV of U.S. Letters Patent No. 5,051,222 issued on September 24, 1991, the specification for which is incorporated by reference herein. Thus, the polyvinyl alcohol or the partially hydrolyzed polyvinyl acetate is prepared first by polymerizing (via a "free radical" polymerization mechanism) polyvinyl acetate having the formula:



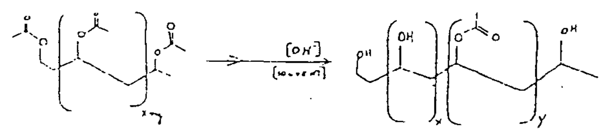
according to the reaction:



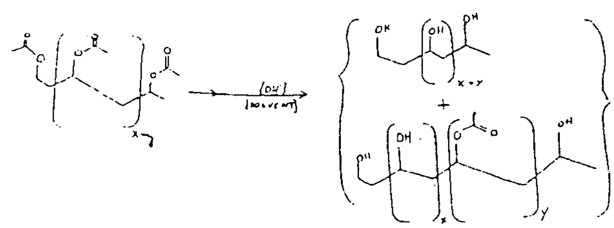
thereby forming a polyvinyl acetate wherein **x + y** are such that the number average molecular weight of the final product is between 5,000 and 67,000 The resulting polyvinyl acetate having the formula:



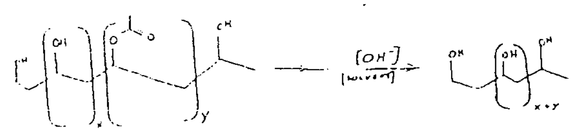
is then hydrolyzed first to form a partially hydrolyzed polyvinyl acetate according to the reaction,



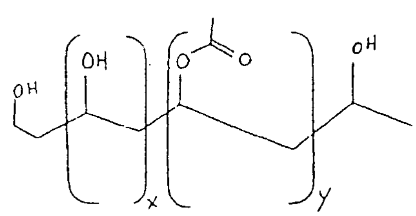
or a mixture of polyvinyl alcohol and partially hydrolyzed polyvinyl acetate according to the reaction:



[0018] If desired, the partially hydrolyzed polyvinyl acetate may be further hydrolyzed to form polyvinyl alcohol with very few acetyl groups present (thereby forming, for example, 99% hydrolyzed polyvinyl acetate) according to the reaction:

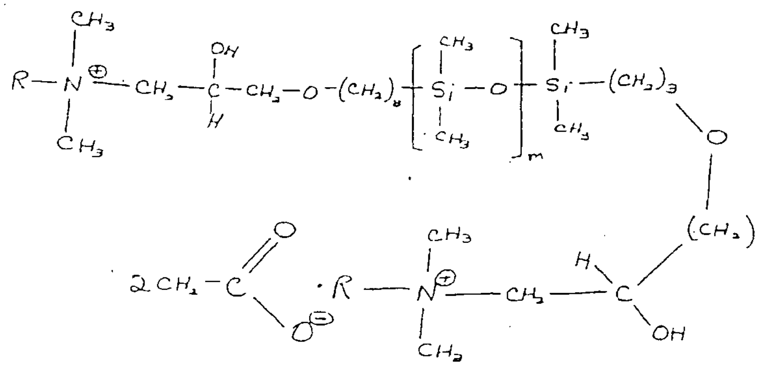


[0019] In any event, the ratio of acetyl moieties to hydroxyl moieties is less than about 1:3 in the structure:



and **x** and **y** are defined whereby **x + y** gives rise to a polymer that has a number average molecular weight of between about 5,000 and 67,000.

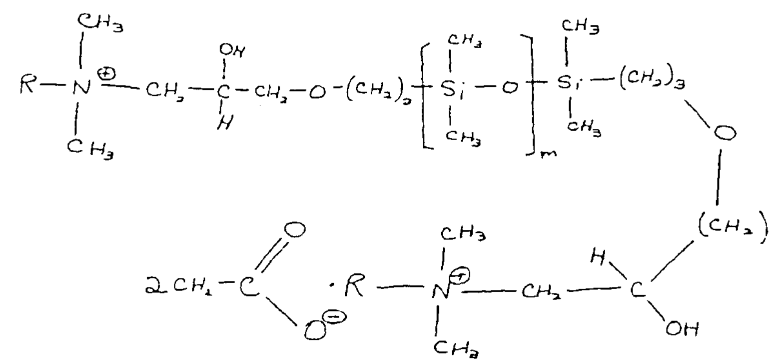
[0020] When creating particles having 10% candelilla wax and 10% fragrance (making up, for example, a fabric softener containing 0.72% fragrance) using surfactants containing both hydrolyzed polyvinyl acetate (99% hydrolzyed) and either the quaternary ammonium polysilane derivatives defined according to the structure:



wherein **R** is the moiety:

$$CH_3\text{-}[CH_2]_x\text{-}$$

and wherein **m** is an integer of from 10 up to 100 and wherein **x** is an integer of from 10 up to 100, or the cationic modified starch, RediBOND® 5320 (trademark of National Starch Inc. of Bridgewater, New Jersey), the following table shows the differences in fragrance intensity on a scale of 1-10:

Surfactant Component in Microparticle	Fragrance Intensity on a Scale of 1 to 10
1% quaternary ammonium polysilane derivative (SILQUAT®) and 2% 99% hydrolyzed polyvinyl acetate	8
1% quaternary ammonium polysilane derivative having the structure:  wherein R is the moiety: $CH_3-\left[CH_2\right]_x-$ and wherein m is an integer of from 10 up to 100 and wherein x is an integer of from 10 up to 100 and 4% 99% hydrolyzed polyvinyl acetate.	4
1% cationic modified starch and 2% 99% hydrolyzed polyvinyl acetate.	7
1% cationic modified starch and 4% 99% hydrolyzed polyvinyl acetate.	6

[0021] Our invention is also directed to a process for fragrancing a perfumable material having a substantially solid surface, such as hair, fabric and mammalian skin, comprising the step of contacting said solid surface of said perfumable material with at least one particle as defined, supra. When carrying out this process, the intensity of fragrancing, Δ**A**, is governed by the algorithm:

$$\Delta A = \alpha \sum_{k=1}^{P} \sum_{j=1}^{Q} \sum_{i=1}^{n} \beta_k M_{0j} \left(1 - e^{-\frac{3 D_j \theta^2}{2 R_i^2}}\right)$$

wherein α is a constant, β_k is the individual and multiple threshold values of the **Q** components of the fragrance material within the microparticle being controllably released (the number of threshold values is "**P**" since not only are individual components measured for their thresholds, but pairs and triplets of fragrance materials are measured for their thresholds also); the symbol M_{0j} is the initial number of gram moles of one of **Q** fragrance components in the particle; D_j is the diffusivity of each of **Q** fragrance components in the particle; θ is the time during which the particle diffusably and controllably releases the fragrance to the solid surface and environment surrounding the particle; and R_i is the radius

of **n** particles. The aroma intensity created from one particle is shown by the equation:

$$\Delta A_P = \alpha \sum_{K=1}^{P} \sum_{j=1}^{Q} \beta_K M_{oj} \left(1 - e^{-\frac{3 D_j \theta^2}{2 R^2}}\right).$$

[0022] The aroma intensity created by **n** particles having an average radius $\bar{R}$ is shown by the equation:

$$\Delta A_{EST.} = \alpha n \sum_{K=1}^{P} \sum_{j=1}^{Q} M_{oj} \left(1 - e^{-\frac{3 D_j \theta^2}{2 \bar{R}^2}}\right).$$

[0023] The foregoing equations are derived using the differential equations:

$$\left(\frac{\partial C_j}{\partial \theta}\right) = D_j \left(\frac{\partial^2 C_j}{\partial x^2} + \frac{\partial^2 C_j}{\partial y^2} + \frac{\partial^2 C_j}{\partial z^2}\right)$$

and

$$\left(\frac{\partial M_j}{\partial \theta}\right) = -\frac{D_j M_j A_j \theta}{V_i R_i}.$$

[0024] The rate of change with respect to time of the aromatization, Δ**A** is shown by the equation:

$$\frac{d\,\Delta A}{d\theta} = \sum_{K=1}^{P} \sum_{j=1}^{Q} \sum_{i=1}^{n} \beta_K \left\{\frac{M_{oj} D_j \theta \alpha}{R_i^2}\right\} \left[4 e^{-\frac{3}{2}\frac{D_j \theta^2}{R_i^2}} - 3 e^{-\frac{3 D_j \theta^2}{R^2}}\right]$$

[0025] In the foregoing compositions, various grades of partially hydrolyzed and substantially fully hydrolyzed forms of hydrolyzed polyvinyl acetate can be used, to wit:

Brand of Hydrolyzed Polyvinyl Acetate (Manufactured by Hoechst A.g., D6230 Frankfurt am Main, Germany)	Number Average Molecular Weight	% Hydrolyzed
MOWIOL® 10-74 (trademark of Hoechst A.g.)	20,000	74
MOWIOL® 4-80	24,000	80
MOWIOL® 3-83	14,000	83
MOWIOL® 5-88	37,000	88
MOWIOL® 8-88	67,000	88
MOWIOL® 3-98	16,000	98

(continued)

Brand of Hydrolyzed Polyvinyl Acetate (Manufactured by Hoechst A.g., D6230 Frankfurt am Main, Germany)	Number Average Molecular Weight	% Hydrolyzed
MOWIOL® 4-98	27,000	98
MOWIOL® 6-98	47,000	98
MOWIOL® 10-98	61,000	98
MOWIOL® GE 4-86	62,000	86

[0026] Additional equations concerning the diffusion of the active or bioactive product from the particulate compositions of our invention are derived using the teachings of Peppas, et al, *Journal of Controlled Release,* Volume 40 (1996), pages 245-250 and entitled "Controlled release of fragrances from polymers I. Thermodynamic analysis" and from the text entitled *DIFFUSION IN POLYMERS* edited by P. Neogi, published 1996 by Marcel Dekker, Inc at pages 165-169 (chapter by Duda and Zielinski entitled "FREE-VOLUME THEORY" and the subchapter entitled "Multicomponent Diffusion". Each of the foregoing references is incorporated by reference herein.

[0027] Our invention is also directed to a process for preparing hydrophobic active ingredient- or bioactive ingredient-containing compositions as defined, supra, comprising the steps of:

(i) intimately admixing at least one hydrophobic active ingredient or bioactive ingredient material with at least one hydrophobic polymer and/or at least one hydrophobic wax to form a first mixture at a temperature greater than or equal to the melting point of said polymer or said wax or, in the case of mixtures, the melting point of the highest melting polymer or wax in the mixture;

(ii) intimately admixing a surfactant (as defined, supra) with an aqueous composition comprising water (for example, a mixture of sodium chloride and water or a mixture of propylene glycol and water or water itself) to form a second mixture which is an aqueous solution (for example, a solution of sodium chloride in water or a solution of propylene glycol in water);

(iii) blending said first mixture and said second mixture at a temperature in the range of from about 60°C up to the boiling point at atmospheric pressure of the aqueous composition (for example, water boiling at 100°C or a mixture of water and propylene glycol boiling at 120°C) whereby a microemulsion is formed; and

(iv) causing the hydrophobic active ingredient- or bioactive ingredient (e.g., perfume)-containing composition in the solid phase to form as an aqueous suspension of solid phase particles (as by cooling to 25°C)

wherein the weight percent of active ingredient or bioactive ingredient (e.g., fragrance composition or aroma chemical) for forming said first mixture is in the range of from about 5% up to about 60% by weight of said first mixture; wherein the weight percent of surfactant in the second mixture is from about 0.01% up to about 5% by weight of said second mixture. In fact, the cooling step, cooling the aqueous suspension, can be carried out at a temperature of from about 10°C up to about 30°C.

[0028] The foregoing process is carried out preferably using a homogenizer and/or a rotor/stator high shear mixer. Examples of a homogenizer useful in the practice of this aspect of our invention are laboratory homogenizer models 15MR and 31MR manufactured by APV Gaulin Inc. of 44 Garden Street, Everett, Massachusetts 02149 Examples of rotor/stator high shear mixers are the high shear in-line mixers manufactured by Silverson Machines, Inc., P.O. Box 589, 355 Chestnut Street, East Long Meadow, Massachusetts 01028 and by the Scott Process Equipment Corporation, P.O. Box 619, Sparta, New Jersey 07871. The aforementioned homogenizers and rotor/stator high shear mixers can be used in conjunction with one another, with the rotor/stator high shear mixers being used first and then in order to bring the particle size down further, the resulting emulsion is then further homogenized using the homogenizers such as laboratory homogenizers, models 15MR and 31MR.

[0029] The details of the aforementioned homogenizers and rotor/stator high shear mixers are set forth in the "DETAILED DESCRIPTION OF THE DRAWINGS" section, infra.

[0030] Our invention is also intended to cover a process for preparing the hydrophobic active or bioactive ingredient-containing compositions discussed, supra (e g., perfume compositions), comprising the steps of:

(i) intimately admixing at least one hydrophobic active or bioactive material (e g , perfume composition) with (a) at least one hydrophobic polymer and/or at least one hydrophobic wax and (b) at least one surfactant to form a

first single liquid phase mixture at a temperature greater than or equal to the melting point of said polymer or said wax or, in the case of mixtures, the melting point of the highest melting polymer or wax in the mixture;

(ii) blending said first single liquid phase mixture with an aqueous composition comprising water (for example, water itself or a mixture of propylene glycol and water or a mixture of sodium chloride and water, for example, a 5% sodium chloride solution or a 20% aqueous propylene glycol solution) whereby a microemulsion is formed; and

(iii) causing the hydrophobic active or bioactive ingredient-containing composition (e.g., a perfume-containing composition or an aroma chemical-containing composition) in the solid phase to form as an aqueous suspension of solid phase particles (for example. cooling the resulting suspension to a temperature in the range of from about 10°C up to about 30°C)

wherein the weight percent of active ingredient or bioactive ingredient for forming the first mixture is in the range of from about 5% up to about 60% by weight of said first mixture; and wherein the weight percent of surfactant in the first mixture is from about 0 01% up to about 5% by weight of the first mixture.

[0031] Again, as stated, supra, with respect to the first-described process for preparing hydrophobic active ingredient- or bioactive ingredient-containing compositions of our invention, the blending step is carried out using a homogenizer and/or a rotor/stator high shear mixture as described in detail, supra, and as exemplified in detail, supra, and as described in detail in the DETAILED DESCRIPTION OF THE DRAWINGS section, infra.

[0032] Our invention is also directed to apparatus for carrying out the aforementioned processes for preparing the hydrophobic active ingredient- or bioactive ingredient-containing compositions. This apparatus comprises:

(i) means for intimately admixing at least one hydrophobic active ingredient- or bioactive ingredient-containing material with at least one hydrophobic polymer or at least one hydrophobic wax to form a first single liquid phase mixture at a temperature greater than or equal to the melting point of said polymer or said wax or, in the case of mixtures, the highest melting component of the mixture;

(ii) means for intimately admixing a surfactant with an aqueous composition comprising water to form a second mixture which is an aqueous solution (for example, using a homogenizer or rotor/stator high shear mixer);

(iii) means for blending said first mixture and said second mixture at a temperature of between 60°C and the boiling point of the aqueous composition at atmospheric pressure whereby a microemulsion is formed (for example, using the homogenizer and/or the rotor/stator high shear mixer as described, supra); and

(iv) means for causing the hydrophobic active ingredient- or bioactive ingredient-containing composition in the solid phase to form as an aqueous suspension of solid phase particles (for example, using cooling means to cool the mixture to 10-30°C, for example, using apparatus equipped with cooling coils).

[0033] Additional apparatus for preparing the hydrophobic active or bioactive ingredient-containing compositions of our invention comprise:

(i) means for intimately admixing at least one hydrophobic active ingredient- or bioactive ingredient-containing composition with (a) at least one hydrophobic polymer and/or at least one hydrophobic wax and (b) at least one surfactant to form a first single liquid phase mixture at a temperature greater than or equal to the melting point of said polymer or said wax or, in the case of mixtures, the melting point of the highest melting of the materials in the mixture;

(ii) means for blending said first single liquid phase mixture with an aqueous composition comprising water whereby a microemulsion is formed (for example, using the homogenizer and/or the rotor/stator high shear mixer as described, supra); and

(iii) means for causing the hydrophobic active ingredient- or bioactive ingredient-containing composition in the solid phase to form as an aqueous suspension of solid phase particles (for example, cooling coils to cool the suspension to a temperature of between 10°C and 30°C).

[0034] Our invention is also directed to fragrance diffusion evaluation apparatus for testing the diffusivity, odor character and odor intensity of a fragrance material and a process for using same. The apparatus of our invention includes at least one hollow container having a test sample suspended therein, air flow means for passing air through the inside

of the container means and an exit port for the air flow where the intensity and aroma character are measured as a function of time and as a function of temperature The sample is weighed initially and at time intervals while the air is flowing through the cylinder(s) at a fixed or variable rate.

[0035] More specifically, this aspect of our invention is directed to apparatus located at an **X-Y-Z** three-space for simultaneously testing the diffusivity, odor character and odor intensity of a fragrance material selected from the group consisting of one or more aroma chemicals and one or more fragrance compositions comprising

(a) hollow vertically disposed substantially cylindrical container means having:

(i) a central axis parallel to the **Z** axis;

(ii) an inner void,

(iii) a substantially circular non-interrupted base composed of a first solid nonporous substance located in a first **X-Y** plane, said base being substantially perpendicular to said **Z** axis and having an inner void side and an outside;

(iv) vertically surrounding said inner void and parallel to and substantially equidistant from said **Z** axis, a continuous cylindrical sidewall composed of a second nonporous substance having an inner void side and an outside, having a top circular rim located in a second **X-Y** plane, an upper midportion, a lower midportion and a bottom rim located in said first **X-Y** plane, said bottom rim being fixedly sealed along its entire circumference to the circumference of said base, said sidewall having a first orifice therethrough located in the lower midportion thereof in a third **X-Y** plane and a second orifice therethrough substantially diametrically opposed to said first orifice located in a fourth **X-Y** plane, said fourth **X-Y** plane being located between and substantially parallel to said third **X-Y** plane and said first **X-Y** plane, said sidewall otherwise being non-interrupted;

(v) a top substantially circular cover composed of a third solid nonporous substance and having an inner void side and an outside sealably attached along its entire outer circumference to the circumference of the top circular rim of said sidewall and located in said second **X-Y** plane, having a substantially circular top cover opening having an inner circumference, said inner circumference being substantially equidistant from said **Z** axis, said opening having a diameter of from about 20% up to about 40% of the outside diameter of the top cover;

(b) sealably mounted through said first orifice, a temperature probe having a temperature variation-sensitive end attached to support means, said temperature variation-sensitive end being located within said inner void, said support means seabably inserted through said first orifice and connected to temperature monitoring means outside said container means;

(c) sealably mounted through said second orifice, air flow means for supplying a stream of air into said inner void, said air flow means comprising a supply tube having an open end located within said inner void, being connected to said supply tube and located outside said container means, an air supply source; and

(d) suspension means for suspending a test sample located within said inner void, said suspension means comprising a substantially flexible support fixedly attached at two substantially diametrically opposed attachment locations located in substantially the same **X-Y** plane proximate said second **X-Y** plane, each being proximate the top rim of said sidewall and having attached thereto and suspended therefrom at a location substantially midway between said attachment locations, said test sample initially having absorbed thereon said fragrance material

whereby when the air supply means is engaged thereby causing air to flow through said air flow means, and the temperature monitor is engaged, air at a fixed temperature $\mathbf{T}$ or a variable temperature $\mathbf{T}(\theta)$ will flow at a fixed flow rate $\mathbf{Q}$ or a variable flow rate $\mathbf{Q}(\theta)$ past said sample having an area $\mathbf{A}$ initially holding $\mathbf{G_0}$ grams of fragrance materials in a concentration of $\mathbf{C_0}$ gram-moles per liter for a period of time θ at the end of which time said sample will hold $\mathbf{G_1}$ grams of fragrance material in a concentration $\mathbf{C_1}$ gram-moles per liter during which time θ the odor character and intensity is capable of being measured and determined proximate the intersection of said **Z** axis with said second **X-Y** plane.

[0036] This aspect of our invention is further directed to such apparatus wherein (i) the temperature of the air supplied by the air supply means is controlled and (ii) the temperature probe has programmed feedback means connected to the air supply means.

[0037] This aspect of our invention is further directed to such apparatus wherein the sidewall has a multiplicity of additional orifices at different locations, each of which has sealably mounted therethrough a temperature probe.
[0038] Such temperature probes each may have programmed feedback means connected to the air supply means and the temperature of the air supplied by the air supply means is controlled.
[0039] This aspect of our invention is further directed to measurement of the odor character and intensity using an electronic aroma testing device.
[0040] This aspect of our invention is further directed to a process for simultaneously testing the diffusivity, odor character and odor intensity of a fragrance material selected from the group consisting of one or more aroma chemicals and one or more fragrance compositions comprising the steps of:

(a) providing the apparatus as defined above;

(b) suspending a test sample containing a fragrance material in the suspension means of the apparatus described above; and

(c) engaging the air supply means and temperature monitor means

whereby when the air supply means is engaged to thereby causing air to flow through the air flow means and the temperature monitor is engaged, air at a fixed temperature **T** or a variable temperature **T**(θ) will flow at a fixed flow rate **Q** or a variable flow rate **Q**(θ) past said sample having an area **A** initially holding $\mathbf{G_0}$ grams of fragrance materials in a concentration of $\mathbf{C_0}$ gram-moles per liter for a period of time θ at the end of which time θ said sample will hold $\mathbf{G_1}$ grams of fragrance material in a concentration $\mathbf{C_1}$ gram-moles per liter during which time θ the odor character and intensity is capable of being measured and determined proximate the intersection of said **Z** axis with said second **X-Y** plane.
[0041] This aspect of our invention is further directed to such apparatus as described above wherein the entirety of the inner void side of the base, the sidewall and the top cover is covered with an absorption-preventing substance such as aluminum foil in order to prevent absorption of the fragrance material into the base, the sidewall and the top cover.
[0042] This aspect of our invention is further directed to apparatus consisting of two separate cylinders operated simultaneously in addition; to simply using one single cylinder.
[0043] This aspect of our invention is further more specifically directed to apparatus wherein the container means has:

(i) a height of between about 50 and about 75 cm;

(ii) a radius of between about 15 and about 30 cm;

(iii) a volume of between 0.1 and about 0.2 m^3;

(iv) a vertical distance of temperature probe to base of between about 10 and about 30 cm;

(v) a vertical distance of air flow means from base being from about 3 up to about 10 cm; and

(vi) a diameter of inner opening in top cover being from about 15 up to about 30 cm.

[0044] This aspect of our invention is further directed to the processes described above wherein the internal pressure of the inner void is maintained at from about 0.5 up to about 2 psig and the air flow rate is maintained in the range of from about 900 up to about 1,000 ml per minute.

BRIEF DESCRIPTION OF THE DRAWINGS

[0045] Figure 1A is a diagram of magnified (2,000x) cloth treated by fabric softener without the use of the microparticulate compositions of our invention.
[0046] Figure 1A-I is a photograph (magnified 2,000x) of cloth treated by fabric softener without the use of the microparticulate composition of our invention.
[0047] Figure 1B is a diagram of magnified fabric (2,000x) washed with wax microparticles containing fragrance of our invention.
[0048] Figure 1B-I is a magnified photograph (2,000x) of fabric (towels) washed with encapsulated fragrance in wax microparticles of our invention.

[0049] Figure 2A is a diagram of a magnified (2,000x) strand of hair washed with shampoo without the use of the microparticulate composition of our invention

[0050] Figure 2A(I) is a magnified (2.000x) photograph of a strand of hair washed with shampoo without the use of the microparticulate composition of our invention

[0051] Figure 2B is a diagram of a magnified (2,000x) strand of hair washed with shampoo containing encapsulated fragrance in wax microparticles of our invention

[0052] Figure 2B(I) is a magnified (2,000x) photograph of a strand of hair treated with shampoo and the encapsulated fragrance in wax microparticles of our invention

[0053] Figure 2C is a diagram of a magnified strand of hair (1,500x) washed with conditioner without the use of the microparticulate composition of our invention.

[0054] Figure 2C(I) is a photograph (magnified 1,500x) of a strand of hair washed with conditioner but without the use of the microparticulate composition of our invention.

[0055] Figure 2D is a diagram of a magnified (1,500x) strand of hair washed with conditioner containing the wax microparticles of our invention.

[0056] Figure 2D(I) is a photograph of a magnified (1,500x) strand of hair washed with conditioner containing wax microparticles of our invention.

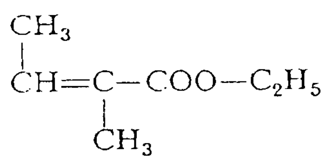
[0057] Figure 3A is a schematic representation of a typical fabric consisting of interwoven bundles which are made up of intertwined fibers. Illustrated is a microparticle entrapped in the pores between the bundles.

[0058] Figure 3B is a schematic representation of a typical fabric consisting of interwoven bundles which are made up of intertwined fibers showing microparticle entrapment in the pores between the bundles as well as direct adhesion through physical forces between a microparticle and a bundle The microparticles are depicted as black dots

[0059] Figure 4A is a cutaway side elevation view of apparatus used to carry out the IFF permeation test in order to determine the permeability of fragrances through a given polymer in the presence or in the absence of surfactant.

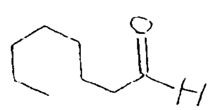
[0060] Figure 4B is a perspective view of the permeation test (diffusion cell) of Figure 4A.

[0061] Figure 5A is a graph indicating the permeability of candelilla wax to the aroma chemicals, ethyl tiglate having the structure:



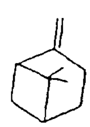
,

aldehyde C-8 having the structure:



,

and β-pinene having the structure:



.

[0062] Figure 5B is another graph showing the permeability of carnauba wax to the aroma chemicals: ethyl tiglate, aldehyde C-8 and β-pinene.

[0063] Figure 5C is another graph showing the permeability of carnauba wax to the aroma chemicals: ethyl tiglate and β-pinene, using non-entrapped ethyl tiglate and β-pinene as controls.

[0064] Figure 5D is a graph showing the permeability of polyethylene wax (molecular weight 500) to the aroma

chemicals: ethyl tiglate, aldehyde C-8 and β-pinene determined by the apparatus of Figures 4A and 4B.

[0065] Figure 5E is a graph showing the permeability of β-pinene through the waxes: cetyl palmitate (CUTINA® wax), carnauba wax, polyethylene wax (molecular weight 500), candelilla wax and a control.

[0066] Figure 5E(A) is an enlargement of that part of Figure 5E wherein the weight loss of product tested is between about zero and about 1 $\frac{mg \cdot mm}{cm^2}$.

[0067] Figure 5F is a graph showing the permeability of ethyl tiglate through the waxes: cetyl palmitate (CUTINA® wax), carnauba wax, polyethylene wax, candelilla wax and a control.

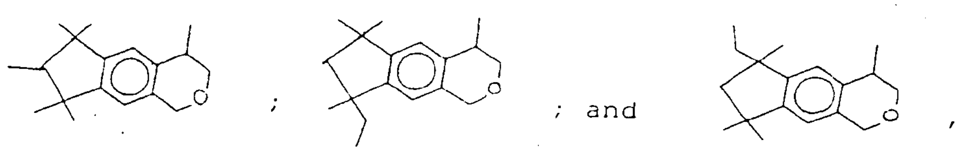
[0068] Figure 5G is a graph showing the permeability of hydroxypropyl celullose to the aroma chemicals: β-pinene and ethyl tiglate and showing the use of a control (without the use of the control release polymer or wax of our invention).

[0069] Figure 5H is a graph showing the permeability of polyvinyl alcohol to the aroma chemicals: ethyl tiglate and β-pinene and also showing the use of a control without the use of the polyvinyl alcohol.

[0070] Figure 6A sets forth a bar graph showing percent geraniol substantivity on cotton fabric swatches for neat geraniol and for geraniol encapsulated in candelilla wax microparticles. The substantivity is plotted on a logarithmic scale.

[0071] Figure 6B sets forth geraniol substantivity on polyester fabric swatches for neat geraniol and for geraniol encapsulated in candelilla wax microparticles. The substantivity is plotted on a logarithmic scale.

[0072] Figure 6C sets forth the substantivity of GALAXOLIDE® (trademark of International Flavors & Fragrances Inc. of New York, NY), a mixture of compounds having the structures:

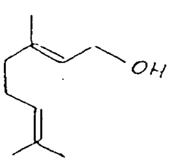


on cotton swatches for the neat GALAXOLIDE® and for GALAXOLIDE® encapsulated in candelilla wax particles.

[0073] Figure 6D sets forth the substantivity of GALAXOLIDE® on polyester fabric swatches for the neat GALAXOLIDE® and for GALAXOLIDE® encapsulated in candelilla wax microparticles

[0074] Figure 7A is a graph showing the sustained release of GALAXOLIDE® over two days as the neat aroma chemical and when encapsulated in candelilla wax microparticles.

[0075] Figure 7B sets forth a graph showing the release of geraniol having the structure:



from the microparticle slurry which was applied to brown hair by washing in water. The release from the slurry includes contributions from both the neat and encapsulated aroma chemical, geraniol.

[0076] Figure 8A is a graph showing odor intensity versus time for fragrance **S** in combination with encapsulated fragrance 361 and fragrance **S** in combination with unencapsulated fragrance 361.

[0077] Figure 8B is a graph showing odor diffusivity versus time for mixtures of fragrance **S** with encapsulated fragrance 361 and fragrance **S** with unencapsulated fragrance 361.

[0078] Figure 8C is a graph showing odor intensity versus time for fragrance **S** in combination with encapsulated fragrance 885 and fragrance **S** with unencapsulated fragrance 885.

[0079] Figure 8D is a graph showing odor diffusivity versus time for fragrance **S** with encapsulated fragrance 885 and fragrance **S** with unencapsulated fragrance 885.

[0080] Figure 8E is a graph showing odor intensity versus time for fragrance **S** with encapsulated fragrance 075 and fragrance **S** with unencapsulated fragrance 075, the encapsulation being in carnauba wax.

[0081] Figure 8F is a graph showing odor diffusivity versus time for fragrance **S** with encapsulated fragrance 075 and fragrance **S** with unencapsulated fragrance 075, the encapsulation being in carnauba wax.

[0082] Figure 8G is a graph showing odor intensity versus time for fragrance **S** and for fragrance **S** with encapsulated

fragrance 361 in carnauba wax.

[0083] Figure 8H is a graph showing odor diffusivity versus time for fragrance S alone and for fragrance S in combination with encapsulated fragrance 361 in carnauba wax.

[0084] Figure 8I is a graph showing odor intensity versus time for fragrance S and for fragrance S in combination with encapsulated fragrance 885 in carnauba wax

[0085] Figure 8J is a graph showing odor diffusivity versus time for fragrance S alone and for fragrance S in combination with encapsulated fragrance 885 in carnauba wax.

[0086] Figure 8K is a graph showing odor intensity versus time for fragrance S taken alone and for fragrance S in combination with encapsulated fragrance 075.

[0087] Figure 8L is a graph showing odor diffusivity versus time for fragrance S taken alone and for fragrance S in combination with encapsulated fragrance 075 in carnauba wax

[0088] Figure 9A is a schematic diagram of a cutaway side elevation view of a microparticle of our invention showing substantially hydrophilic surfactant substantially entirely coated and fixedly bonded to the entirety of the outer surface of a single phase solid solution in the form of a continuous submicron layer of surfactant.

[0089] Figure 9B shows a solid solution-microparticle of our invention wherein the substantially hydrophilic surfactant is located proximate to and immediately, substantially beneath the entirety of the outer surface of the solid solution and substantially within the internal matrix volume.

[0090] Figure 9C shows a particle of our invention wherein the substantially hydrophilic surfactant is both (a) substantially entirely coated on and fixedly bonded to the entirety of said outer surface of said single phase solid solution in the form of a continuous submicron layer of surfactant and (b) located proximate to and immediately, substantially beneath the entirety of said outer surface of said solid solution and substantially within said internal matrix volume.

[0091] Figure 10A is a schematic diagram showing the side view of a diffusivity testing apparatus for testing the diffusivity of entrapped and nonentrapped fragrance materials including aroma chemicals and fragrance compositions.

[0092] Figure 10B is the top view of the apparatus of Figure 10A.

[0093] Figure 11A is a schematic perspective view of the first stage of the operation of a rotor/stator high shear mixer, wherein the high speed rotation of the rotor blades within the precision machined mixing workhead exerts a powerful suction drawing liquid and solid materials into the rotor/stator assembly.

[0094] Figure 11B is a schematic perspective diagram of stage two of the operation of a rotor/stator high shear mixer used in the processes and apparatus of our invention where centrifical force drives materials towards the periphery of the workhead where they are subjected to a milling action in the precision machined clearance between the ends of the rotor blades and the inner wall of the stator.

[0095] Figure 11C is a schematic perspective diagram of the operation of the third stage of a rotor/stator high shear mixer useful in the apparatus of our invention and in carrying out the processes of our invention, wherein the second stage is followed by intense hydraulic shear as the materials are forced, at high velocity, out through the perforations in the stator, then through the machine outlet and along the pipework, while at the same time, fresh materials are continually drawn into the workhead, maintaining the mixing and pumping cycle.

[0096] Figure 11D is a schematic side view of the homogenizing equipment assembly for carrying out the blending step of the processes of our invention and as part of the apparatus of our invention.

[0097] Figure 11E is a schematic cutaway, side elevation view of a single-stage homogenizing valve assembly for the homogenizing part of the apparatus of our invention.

[0098] Figure 11F is a cutaway side elevation view of a two-stage homogenizing valve assembly for the homogenizing apparatus for the blending step of the processes of our invention and for the apparatus of our invention

[0099] Figure 11G is a schematic side elevation view of a rotor/stator mixing assembly useful in the apparatus of our invention and in carrying out the blending step of the processes of our invention.

[0100] Figure 12A sets forth a block flow diagram showing the process steps of our invention in preparing particulate compositions of our invention. Figure 12A also shows in schematic form the apparatus of our invention.

[0101] Figure 12B is a schematic block flow diagram setting forth the process steps of our invention for the process useful in preparing the compositions of our invention. Figure 12B also sets forth in schematic form the apparatus of our invention.

[0102] Figure 12C shows a schematic diagram of the apparatus and process steps of Figure 12A with an additional schematic representation of the utilization of an electronic program controller (e.g., computer system) whereby market demand information and the like can be utilized to cause automatic alterations in the process variables of the process of our invention where ingredients are admixed, blended, heated and cooled.

[0103] Figure 12D shows a schematic diagram of the apparatus of Figure 12B with an additional schematic representation of the utilization of an electronic program controller (e g., computer system) whereby market demand information and the like can be utilized to cause automatic alterations and adjustments in the process variables (e g. blending, heating, ratio of ingredients and cooling as well as flow rates) in the process of our invention and in the apparatus of our invention

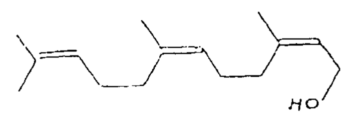
[0104] Figure 13 is a schematic flow diagram showing the processing in schematic form of wax microparticles containing active or bioactive ingredients of our invention

[0105] Figure 14 is a graph showing log vapor pressure versus accumulated sum of fragrance ingredients for three different fragrances: fragrance A-1, fragrance A-2 and fragrance A-3. Each of the fragrances falls within the same graph.

[0106] Figure 15 is a graph showing weight loss (weight percent) normalized versus time for three different groups of ingredients: low vapor pressure materials, high vapor materials and mixtures of low and high vapor pressure materials.

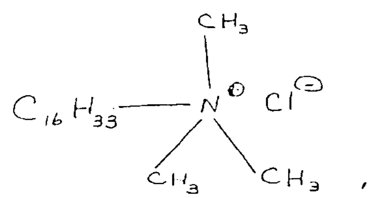
[0107] Figure 16 is a graph showing weight loss rate (weight percent per minute) versus weight percent in the formula of fragrance A-3, a high vapor pressure fragrance, for both the fragrance diffusion evaluation system (as set forth in Figures 10A and 10B) and for the prior art thermal gravimetric analysis system.

[0108] Figure 17 is a graph showing aroma chemical loading efficiencies for the chemicals: benzyl alcohol, geraniol, farnesol having the structure:



and GALAXOLIDE® (registered trademark of International Flavors & Fragrances Inc.) versus $\log_{10}P$ wherein **P** is the octanol-water partition coefficient for the aroma chemicals. The microparticles which are the subject of this graph are candelilla wax microparticles.

[0109] Figure 18A sets forth a particle-size distribution for particles evolving out of the rotor/stator mixer using a composition containing 1.2% cetyl trimethyl ammonium chloride having the structure:



10% candelilla wax and 10% fragrance P-50448. The graph shows volume percent versus particle diameter.

[0110] Figure 18B is a graph of volume percent versus particle diameter for particles evolving out of the homogenizer blending apparatus, which particles contain 5% cetyl trimethyl ammonium chloride, 10% candelilla wax and 10% fragrance IB-X-016.

[0111] Figure 19 is a graph of air flow (ml/minute) versus time (minute) showing a system of our invention using two cylinders as set forth in Figure 10B described, supra, and described in detail, infra, with the two cylinders being connected in parallel.

DETAILED DESCRIPTION OF THE DRAWINGS

[0112] Referring to Figure 1A and Figure 1A-I, reference numeral 10 indicates the fiber itself and reference numeral 11 indicates the space between the fibers.

[0113] Referring to Figure 1B and Figure 1B(I), reference numeral 12 refers to the wax microparticles located on the surface of the fabric.

[0114] Referring to Figure 2A and Figure 2A(I), reference numeral 20 refers to the hair strand itself

[0115] Referring to Figures 2B and 2B(I), reference numeral 21 refers to the wax microparticles located on the surface of the hair strands.

[0116] Referring to Figures 2C and 2C(I(), reference numeral 23 refers to the strand of hair washed with a conditioner.

[0117] Referring to Figures 2D and 2D(I), reference numeral 24 refers to the wax microparticles located on the surface of the hair strands.

[0118] Referring to Figure 3A, the bundles of fibers are referred to by reference numerals 30a and 30b. The microparticle entrapped in the pores between the bundles is referred to by reference numeral 32. The space between the

bundles is referred to by reference numeral 31. Figure 3A is an enlargement of section 3A of Figure 3B

[0119] Referring to Figure 3B, the fiber bundles are shown by reference numeral 33. The microparticle entrapped between the bundles is shown by reference numeral 36 being entrapped in space 37. The fiber bundles are shown by reference numerals 33a and 33b and, in addition, reference numerals 34 and 34b. The microparticle directly adhered through physical forces onto a bundle is shown by reference numeral 35.

[0120] Referring to Figures 4A and 4B, fluid 46 is located in jar 44. Jar 44 has sidearm 45. The fluid 46 reaches fluid level 47. Directly in line with fluid level 47 is membrane 41. The diffusion membrane 41 is held in place with flanges 43 and jar lip 42, using bolts 401a and 401b which secures the flange in place. The sidearm 45 is closed using closure 49. The permeability apparatus is shown using reference numeral 40. The IFF permeability test is based on the use of the apparatus of Figures 4A and 4B. The weight of membrane 41 is taken, initially, before being placed within the flange 43 and the jar lip 42. Substance 46 for which the permeability is to be measured is placed into jar 44 to fluid level 47. The apparatus containing fluid 46 remains in place for a fixed period of time. At the end of that period of time, bolts 401a and 401b as well as 401c are loosened, the flange 43 removed and membrane 41 is removed and weighed, thereby gathering sufficient data to determine the permeability of the particular substance 46. Referring to Figure 5A, reference numeral 52 are data points for aldehyde C-8. Reference numeral 53 shows data points for ethyl tiglate. The **Y** axis measures weight loss and is indicated by reference numeral 54. The **X** axis shows time in minutes and is indicated by reference numeral 55. The weight loss is measured in $\frac{mg \cdot mm}{cm^2}$.

[0121] Reference numeral 51 shows the standard deviation for the data.

[0122] In Figure 5B, reference numeral 503 represents data points for aldehyde C-8. Reference numeral 502 represents data points for β-pinene. Reference numeral 58 represents data points for ethyl tiglate. Reference numeral 501 shows the graph for weight loss versus time for ethyl tiglate, thus showing the permeability of carnauba wax to ethyl tiglate. The **Y** axis for weight loss is indicated by reference numeral 56, and the **X** axis for time is indicated by reference numeral 57. Reference numeral 59 sets forth the standard deviation for the data for ethyl tiglate.

[0123] Referring to Figure 5C, reference numeral 507 is for ethyl tiglate, encapsulated. Reference numeral 508 is for the graph of ethyl tiglate for weight loss versus time, showing the permeability of carnauba wax to ethyl tiglate. Reference numeral 506 shows the data points for β-pinene in carnauba wax. Reference numeral 510 shows data points for ethyl tiglate without being entrapped in any control release system such as carnauba wax. Reference numeral 509 shows data points for the β-pinene control in the absence of carnauba wax. Reference numeral 511 sets forth the graph of unentrapped ethyl tiglate. Reference numeral 512 sets forth the graph of unentrapped β-pinene. The **Y** axis showing weight loss is indicated by reference numeral 505. The **X** axis showing time in minutes is indicated by reference numeral 504.

[0124] Referring to Figure 5D, reference numeral 515 sets forth the data points for ethyl tiglate in the polyethylene (molecular weight 500) wax. Reference numeral 520 is for the graph of time versus weight loss. Reference numeral 516 shows data points for aldehyde C-8 in polyethylene wax. Reference numeral 517 shows data points for β-pinene in polyethylene wax. Reference numeral 519 indicates the graph for aldehyde C-8 in polyethylene wax, showing the permeability of aldehyde C-8 through polyethylene wax. Reference numeral 520 shows the graph for ethyl tiglate in polyethylene wax showing the permeability of ethyl tiglate through polyethylene wax. The **X** axis is indicated by reference numeral 513, and the **Y** axis is indicated by reference numeral 514.

[0125] Referring to Figure 5E, reference numeral 528 refers to cetyl palmitate (CUTINA® wax). Reference numeral 527 refers to data points for carnauba wax. Reference numeral 526 refers to data points for polyethylene wax. Reference numeral 525 refers to data points for candelilla wax. Reference numeral 523 refers to the control for β-pinene without wax. All data points set forth on Figure 5E, except for the control, show the permeability of β-pinene through waxes. The **X** axis is indicated by reference numeral 521, showing time and minutes, and the **Y** axis is indicated by reference numeral 522, showing weight loss in $\frac{mg \cdot mm}{cm^2}$.

[0126] Figure 5E(A) shows that portion of Figure 5E where the weight loss is between zero and 1.4 for the β-pinene contained in the waxes. In Figure 5E(A), reference numeral 533 shows the standard deviation line. The **X** axis is indicated by reference numeral 521, and the **Y** axis is indicated by reference numeral 522 for weight loss. Reference numeral 528 shows data points for cetyl palmitate. Reference numeral 526 shows data points for polyethylene wax (molecular weight 500). Reference numeral 530 sets forth the graph for time versus weight loss for polyethylene wax containing β-pinene. Reference numeral 533a is the standard deviation line for the data points for polyethylene wax containing β-pinene. Reference numeral 529 shows the graph for carnauba wax containing β-pinene and sets forth the permeability of β-pinene through carnauba wax.

[0127] Referring to Figure 5F, reference numeral 538 shows the data points for cetyl palmitate (CUTINA® wax). Reference numeral 540 shows data points for carnauba wax. Reference numeral 541 shows data points for polyethylene (molecular weight 500) wax. Reference numeral 542 shows data points for candelilla wax. Reference numeral 537 shows data points for the control, ethyl tiglate without wax. Reference numeral 536 is the graph showing the evaporation rate for the control, ethyl tiglate without wax. Reference numeral 539 is the graph showing permeability of ethyl tiglate through cetyl palmitate (CUTINA® wax). Reference numeral 543 sets forth the graph showing permea-

bility of ethyl tiglate through carnauba wax. The **X** axis is indicated by reference numeral 534 for time (minutes) . The **Y** axis is indicated by reference numeral 535 for weight loss

$$\left(\frac{mg - mm}{cm^2}\right).$$

[0128] Referring to Figure 5G, reference numeral 562 indicates data points for β-pinene contained in hydroxypropyl cellulose. Reference numeral 564 indicates data points for β-pinene not contained in any polymer, but merely showing the evaporation rate of the β-pinene. Reference numeral 569 shows the standard deviation for the data points for β-pinene without being contained in hydroxypropyl cellulose. Reference numeral 563 shows the data points for ethyl tiglate contained in hydroxypropyl cellulose. Reference numeral 565 sets forth the data points for ethyl tiglate not being contained in any polymer and showing the evaporation rate of ethyl tiglate. Reference numeral 567 sets forth the graph showing the permeability of ethyl tiglate through hydroxypropyl cellulose. Reference numeral 568 sets forth the graph showing the evaporation of ethyl tiglate (without being contained in any polymer). Reference numeral 566 sets forth the graph showing the evaporation of β-pinene without being present in any polymer. The **X** axis is indicated by reference numeral 561 showing time in minutes, and the **Y** axis is indicated by reference numeral 560 showing weight loss in $\frac{mg \cdot mm}{cm^2}$.

[0129] Referring to Figure 5H, reference numeral 552 shows the data points for ethyl tiglate contained in polyvinyl alcohol. Reference numeral 553 sets forth the data points for β-pinene contained in polyvinyl alcohol (99% hydrolyzed polyvinyl acetate). Reference numeral 554 sets forth the data points for ethyl tiglate not contained in polyvinyl alcohol and merely shows the evaporation rate of the ethyl tiglate. Reference numeral 555 sets forth the data points for β-pinene not being contained in any polyvinyl alcohol, but merely showing the evaporation rate of the β-pinene. Reference numeral 557 sets forth the graph showing the evaporation rate of β-pinene not being contained in any polyvinyl alcohol. Reference numeral 559 sets forth the standard deviation for the data points for ethyl tiglate and β-pinene not being contained in any polyvinyl alcohol. The **X** axis for time (minutes) is shown by reference numeral 551. The **Y** axis for weight loss

$$\left(\frac{mg - mm}{cm^2}\right)$$

is shown by reference numeral 560.

[0130] Referring to Figure 6A, percent substantivity is shown on the **Y** axis by reference numeral 60. The bar graph for neat geraniol using a plain water wash is shown by reference numeral 61a. The bar graph for geraniol contained in candelilla wax microparticles is shown by reference numeral 61b for a plain water wash. The bar graph for neat geraniol using a detergent is shown by reference numeral 62a. The bar graph for neat geraniol in candelilla wax microparticles is shown by reference numeral 62b for detergents. The bar graph for fabric softeners for neat geraniol is shown by reference numeral 63a. The bar graph for geraniol contained in candelilla wax microparticles in fabric softeners is shown by reference numeral 63b.

[0131] Referring to Figure 6B, substantivity (percent) is shown on the **Y** axis indicated by reference numeral 64. Reference numeral 65a sets forth the substantivity of neat geraniol in a plain water wash on polyester fabric. Reference numeral 65a' sets forth the standard deviation line for the neat geraniol in a plain water wash. Reference numeral 65b sets forth the use of neat geraniol encapsulated in candelilla wax microparticles for a plain water wash. Reference numeral 65b' sets forth the standard deviation line for the encapsulated geraniol in candelilla wax using a plain water wash. Reference numeral 66a sets forth the use of neat geraniol in a detergent on polyester fabrics. Reference numeral 66b sets forth the use of geraniol encapsulated in candelilla wax microparticles used in a detergent. Reference numeral 67a sets forth the use of neat geraniol in a fabric softener on polyester fabrics. Reference numeral 67b sets forth the use of geraniol encapsulated in candelilla wax microparticles used in a fabric softener on polyester fabrics.

[0132] Referring to Figure 6C, the **Y** axis shows substantivity in terms of percentages and is shown by reference numeral 601. Reference numeral 602a sets forth the use of neat GALAXOLIDE® in a detergent on cotton fabric. Reference numeral 602b sets forth the use of GALMOLIDE® encapsulated in candelilla wax microparticles on cotton fabrics in a detergent. Reference numeral 603a sets forth the use of neat GALAXOLIDE® in a fabric softener on cotton fabrics. Reference numeral 603b sets forth the use of GALAXOLIDE® encapsulated in candelilla wax microparticles in fabric softener on cotton fabrics.

[0133] Referring to Figure 6D, substantivity (percent) is set forth on the Y axis using reference numeral 604. In Figure 6D, reference numeral 605a refers to the use of neat GALAXOLIDE® in a detergent on polyester fabrics. Reference numeral 605b refers to the use of GALAXOLIDE® encapsulated in candelilla wax microparticles in a detergent for use on polyester fabrics. Reference numeral 605b' refers to the standard deviation for the use of GALAXOLIDE® encapsulated in candelilla wax microparticles with a detergent on polyester fabrics. Reference numeral 606a refers to the use of neat GALAXOLIDE® in a fabric softener on polyester fabrics. Reference numeral 606a' refers to the standard deviation of the data of neat GALAXOLIDE® in a fabric softener for use on polyester fabrics. Reference numeral 606b refers to the use of GALAXOLIDE° encapsulated in candelilla wax microparticles in a fabric softener for use on polyester fabrics. Reference numeral 606b' refers to the standard deviation of the data for GALAXOLIDE® encapsulated in candelilla wax microparticles for use with a fabric softener on polyester fabrics.

[0134] Referring to Figure 7A, showing the sustained release of GALAXOLIDE® over a period of two days as the neat aroma chemical and when encapsulated in candelilla wax microparticles. Reference numeral 70 indicates the Y axis. Reference numeral 71 indicates the X axis in time in days. Reference numeral 72 sets forth the graph for the use of the neat GALAXOLIDE®. Reference numeral 72' sets forth the data points for the neat GALAXOLIDE®. Reference numeral 73 sets forth the graph for the GALAXOLIDE® contained in the candelilla wax microparticles. Reference numeral 703 sets forth the data points for the GALAXOLIDE® encapsulated in candelilla wax microparticles. Reference numeral 704 sets forth the standard deviation for the data points for GALAXOLIDE® encapsulated in candelilla wax microparticles.

[0135] Referring to Figure 7B, reference numeral 74 refers to the Y axis which refers to the percent of aroma chemical (geraniol) remaining on brown hair, and reference numeral 75 refers to the X axis indicating time of release in days. Reference numeral 76 sets forth the graph showing the rate of release of neat geraniol. Reference numeral 701 sets forth the data points for the neat geraniol. Reference numeral 702 sets forth the standard deviation for the data points for the neat geraniol. Reference numeral 77 sets forth the graph for the rate of release of geraniol encapsulated in candelilla wax microparticles. Reference numeral 79 sets forth the data points for geraniol encapsulated in candelilla wax microparticles. Reference numeral 78 sets forth the standard deviation for the data points for geraniol encapsulated in candelilla wax microparticles.

[0136] Referring to Figure 8A, the Y axis indicating odor intensity on a scale of 1-10 is indicated by reference numeral 80. The X axis indicating time in hours is indicated by reference numeral 81. Reference numeral 82 refers to the graph of odor intensity versus time for fragrance S in combination with encapsulated fragrance 361. Reference numeral 83 refers to the graph of odor intensity versus time for the combination of fragrance S and encapsulated fragrance 361.

[0137] Referring to Figure 8B, the Y axis is indicated by reference numeral 84 for odor diffusivity on a scale of 1-10. The X axis is indicated by reference numeral 85 for time in hours. Reference numeral 86 indicates the graph of odor diffusivity versus time for fragrance S in combination with encapsulated fragrance 361. Reference numeral 87 indicates the graph of odor diffusivity versus time for fragrance S ir combination with unencapsulated fragrances 361.

[0138] Referring to Figure 8C, reference numeral 801 indicates the graph of odor intensity versus time for fragrance S anc encapsulated fragrance 885. Reference numeral 802 indicates the graph of odor intensity versus time for fragrance S and unencapsulated fragrance 885.

[0139] Referring to Figure 8D, reference numeral 803 shows the graph of odor diffusivity versus time for fragrance S and encapsulated fragrance 885. Reference numeral 804 indicates the graph for odor diffusivity versus time for fragrance S in combination with unencapsulated fragrance 885.

[0140] Referring to Figure 8E, reference numeral 805 indicates the graph of odor intensity versus time for fragrance S and encapsulated fragrance 075. Reference numeral 806 sets forth the graph of odor intensity versus time for fragrance S and unencapsulated fragrance 075.

[0141] Referring to Figure 8F, reference numeral 807 shows the graph of odor diffusivity versus time for fragrance S and encapsulated fragrance 075. Reference numeral 808 sets forth the graph of odor diffusivity versus time for fragrance S and unencapsulated fragrance 075. These graphs suggest that the carrier retains the topnotes during storage and adheres to the hair.

[0142] Referring to Figure 8G, reference numeral 809 sets forth the graph for odor intensity versus time for fragrance S taken alone. Reference numeral 810 sets forth the graph of odor intensity versus time for fragance S taken together with encapsulated fragrance 361.

[0143] Referring to Figure 8H, reference numeral 811 sets forth the graph of odor diffusivity versus time for fragance S taken alone. Reference numeral 812 sets forth the graph of odor diffusivity versus time for fragrance S taken in combination with encapsulated fragrance 361. The system containing the encapsulated fragrance clearly provides an advantage over the neat oil. The observed difference in intensity between these systems is large enough to provide a perceived difference.

[0144] Referring to Figure 8I, the graph indicated by reference numeral 813 is for the odor intensity versus time for fragrance S taken alone. The graph indicated by reference numeral 814 is for odor intensity versus time for fragrance S taken together with encapsulated fragrance 885.

[0145] Referring to Figure 8J, the graph indicated by reference numeral 815 is for odor diffusivity versus time for fragrance S taken alone. The graph indicated by reference numeral 816 is for odor diffusivity versus time for fragrance S taken together with encapsulated fragrance 885.

[0146] Referring to Figure 8K, the graph indicated by reference numeral 817 is for odor intensity versus time for fragrance S taken alone. The graph indicated by reference numeral 818 is for odor intensity versus time for fragrance S taken together with encapsulated fragrance 075.

[0147] Referring to Figure 8L, the graph indicated by reference numeral 819 is for odor diffusivity versus time for fragrance S taken alone. The graph indicated by reference numeral 820 is for odor diffusivity versus time for fragrance S taken together with encapsulated fragrance 075. These graphs show that the volume and the tenacity of encapsulated fragrance 075 is higher than that of fragrance S taken alone up to 5 hours. The encapsulation system may be used to increase substantivity.

[0148] Referring to Figure 9A, the substantially hydrophilic surfactant 93 is substantially, entirely coated on and fixedly bonded to the entirety of the outer surface 95 of the single phase solid solution 91 in the form of a continuous submicron layer of surfactant 92. The particle having surfactant coated thereon in a submicron layer is indicated by reference numeral 90.

[0149] Referring to Figure 9B, the substantially hydrophilic surfactant 903 is located proximate to and immediately substantially beneath the entirety of the outer surface 905 of the solid solution 901 and substantially within the internal matrix volume. The charge on the particle is shown using reference numeral 904, and the particle itself is shown by reference numeral by reference numeral 900.

[0150] Referring to Figure 9C, the substantially hydrophilic surfactant 917/913 is both (a) substantially, entirely coated on and fixedly bonded to the entirety of the outer surface 915 of the single phase solid solution 911 in the form of a continuous submicron layer of surfactant 912 and (b) located proximate to and immediately, substantially beneath the entirety of the outer surface of said solid solution 915 and substantially within the internal matrix volume. The surfactant within the matrix volume is indicated by reference numeral 917. The surfactant within the submicron layer is indicated by reference numeral 913. The particle is indicated by reference numeral 910. The charge on the outer surface 916, the particle, is indicated by reference numeral 914.

[0151] Figures 10A and 10B set forth the fragrance diffusion evaluation system for determining the diffusivity and permeability of fragrance materials and other active and bioactive ingredients used in the practice of our invention. The test sample on blotters indicated by reference numeral 1001 are supported by support 1002 in container 1003 having opening 1004 to the atmosphere. Air flow through line 1010 is supplied from air supply 1005 through tube 1006, having pressure gauge 1007 measuring the air flow. Container 1003 has side wall 1012 through which temperature probe 1009 is located. Temperature probe 1009 is attached to temperature monitor 1008. Container 1003 has base 1001. The overall apparatus is indicated by reference numeral 1000. Figure 10B sets forth a top view of the apparatus of Figure 10A showing the use two tandem chambers 1003a and 1003b. Container 1003a is supplied with air flow through tube 1010a having pressure gauge 1007b in the air flow line. Container 1003b is supplied with air flow through tube 1010b with pressure gauge 1007a in its line. Air supply from location 1005 supplies air through line 1006a having pressure gauge 1007 in the line to measure air flow. The air flow is then split between line 1006b (for air flowing into container 1003a) and line 1006c (for air flowing to container 1003b). Temperature probe 1009a is used for container 1003a, and temperature probe 1009b is used for container 1003b. Temperature probe 1009b is attached to temperature monitor 1008b. Temperature probe 1009a is attached to temperature monitor 1008a. Container 1003a has opening 1004a at the top of same. Container 1003b has opening 1004b at the opening thereof. The overall apparatus having tandem containers for testing purposes is indicated by reference numeral 1000'.

[0152] The system shown in Figures 10A and 10B has as its primary purpose the simultaneous evaluation of an air freshener's performance for its hedonics, intensity, volatile content and weight loss as a function of time in a controlled environment of temperature and air mixing. The fragrance diffusion evaluation system is a midway station between a laboratory system that allows only analytical measurements and a full scale test of odor performance in a specially designed room that allows only sensory testing. The fragrance diffusion evaluation system provides a controlled environment that allows for both sensory and analytical measurements of a fragrance's performance at low cost.

[0153] The fragrance diffusion evaluation system, shown in Figures 10A and 10B, comprises a cylinder having a height of between about 50 and about 75 cm, a radius of between about 15 and 30 cm and a volume of between about 0.1 and 0.2 m^3. The interior is coated with aluminum foil to ensure that no fragrance absorbs into the walls. The air flow is provided by a tube through the side between about 3 and about 10 cm from the bottom extending to the center of the chamber. The temperature is continuously monitored by a gauge located between about 10 and about 30 cm from the bottom. An opening with a diameter of between 15 and 30 cm is at the top of the cylinder to allow air flow and odor intensity testing. The air flow is, on average, between about 900 and 1,000 ml per minute. This air flow replaces the whole volume of the fragrance diffusion evaluation system with fresh air every 2 hours. The air flow through the chamber is constant at a pressure of between about 0.5 and 2 psig.

[0154] Referring to Figure 11A, the high speed rotation of the rotor blades 1106 within the precision machine mixing

workhead exerts a powerful suction at location 1101 drawing liquid and solid materials 1104a into the rotor stator assembly 1100 The rotation is effected at access 1102 The output from the assembly is at location 1103 The workhead is indicated by reference numeral 1105 The overall device is indicated by reference numeral 1100 Referring to Figure 11B, centrifugal force then drives materials 1104a towards the periphery of the workhead where they are subjected to a milling action in the precision machined clearance between the ends of the rotor blades and the inner wall of the stator

[0155] Referring to Figure 11C, stage 2 is followed by intense hydraulic shear as the materials 1104b are forced at high velocity out through the perforations in the stator 1106, then through the machine outlet and along the pipework 1103 At the same time, fresh materials are continually drawn into the workhead at 1101, maintaining the mixing and pumping cycle.

[0156] Referring to Figure 11E, the single-stage homogenizing valve assembly, valve handle 1112a is used to adjust the flow inwardly at location 1113a and outwardly at location 1114a

[0157] Referring to Figure 11F, Figure 11F sets forth a two-stage valve assembly for the homogenizer Valve handle 1111 is used to adjust the first stage, and valve handle 1112 is used to adjust the second stage. The two-stage valve assembly contains seals 1117 and gaps 1115 and 1116. Reference numeral 1113 refers to the inlet to the two-stage valve assembly, and reference numeral 1114 refers to the outlet of the two-stage valve assembly. Reference numeral 1118 refers to the passageway between the inlet 1113 and the outlet 1114. The overall two-stage valve assembly is indicated by reference numeral 1110. Referring to Figure 11D, Figure 11D is the homogenizing equipment assembly. Mixer 1120 containing mixing shaft 1112 is a steam-heated feeder tank. The homogenizing equipment assembly is shown with the two-stage pressure adjustment system wherein the first-stage hand wheel is shown by reference numeral 1111 and the second-stage hand wheel is shown by reference numeral 1112. Pressure gauge 1122 is used to monitor the flow of fluid containing emulsion through a three-way bypass valve to cooling coils 1130 and recycle line 1114. Temperature gauge 1124 monitors the temperature of fluid flowing through line 1113 into the two-stage valve assembly which is attached to gear box 1123. The overall homogenizing equipment assembly is indicated by reference numeral 1110

[0158] Referring to Figure 11G, the rotor/stator mixing assembly, the initial blending operation is carried out in steam-heated feeder tank 1140 equipped with stirrer 1146 Fluid flows through line 1144 into rotor/stator mixing head 1142 controlled through control box 1141. The fluid then flows through line 1144 into three-way valve 1150. The fluid flows through cooling coils 1143 and 1143a The fluid also flows past the three-way valve through recycle lines 1147 and 1147a back into feeder tank 1140. The rotor/stator mixing assembly is indicated by reference numeral 1190.

[0159] Referring to Figure 12A, fragrance material from container 1201 flows through line 1203 controlled by valve 1202. Simultaneously, polymer and/or wax from container 1204 heated using heater 1205 flows through line 1207 controlled by valve 1206 Both fragrance material, polymer and/or wax flowing through lines 1203 and 1207 are blended in mixing tank 1208 which is also equipped with heater 1212 The thus-formed blend passes through line 1219 controlled by valve 1218 into blender 1220 simultaneously with product evolving from mixer 1215. Thus, surfactant from container 1209 flows through line 1214 past control valve 1211, and simultaneously, water or aqueous mixture from container 1210, preheated using heater 1230, flows through line 1213 past control valve 1212 into mixing vessel 1215, also equipped with heater 1231. The surfactant/aqueous mixture is then passed through line 1217 past control valve 1216 into blender 1220 along with product from line 1219. Blender 1220 is a homogenizer and/or rotor/stator high shear mixer. Subsequent to the blending using the homogenizer and/or rotor/stator high shear mixer, product is passed through line 1223 past control valve 1222 into solid phase particle formation vessel 1225 equipped with cooling coils 1224, or apparatus components 1220 and 1225 can be combined into assembly 1190 as shown in Figure 11G or assembly 1110 as shown in Figure 11D. The resulting particulate slurry is then passed through line 1227 past control valve 1226 into a vessel for further utilization.

[0160] Referring to Figure 12B, fragrance material from container 1250 is passed through line 1259 past control valve 1258 into mixer 1260 equipped with heater 1261. Simultaneously, polymer and/or wax from container 1251 equipped with heater 1252 is passed through control line 1256 past valve 1257 into mixer 1260. Simultaneously, surfactant from container 1253 is passed through line 1254 past valve 1255 into mixing vessel 1260. While mixing vessel 1260 is engaged in mixing fragrance material, polymer or wax and surfactant, aqueous composition heated through heater 1265 in container 1264 is then passed through line 1266 past control valve 1267 into blender 1268. Product mixed in container 1260 is passed through line 1262 past control valve 1263 into blender 1268. Blender 1268 can be a homogenizer and/or a rotor/stator high shear mixer. The resulting product is then passed through line 1269 past valve 1270 into solid phase particle formation vessel 1271 equipped with cooling coils 1272. In the alternative, apparatus components 1271 and 1268 can be combined into assemblies 1190, as shown in Figure 11G, or 1110, as shown in Figure 11D. The resulting product containing solid particulate particles having continuous surfaces is then passed through line 1273 past valve 1274 into container 1275 for utilization of the slurry.

[0161] The apparatus of Figure 12A can be used in conjunction with electronic program controller 1300 as shown in Figure 12C. Electronic program controller 1300 uses marketing input information from source 1299 via control line 1299c feeding into the electronic program controller 1300 and controlling the apparatus as illustrated in Figures 12A

and 12C via control lines. Thus, the apparatus shown in schematic diagram in Figure 12A is also shown in schematic form in Figure 12C as associated with the electronic program controller (computer mechanism) via control lines.

[0162] More specifically, the control of fragrance material from container 1201 through line 1203 past control valve 1202 is controlled via control line 1202c. By the same token, flow of polymer and/or wax from container 1204 through line 1207 past valve 1206 is controlled through control line 1206c. The rate of heating and amount of heat energy into container 1204 using heater 1205 is controlled through control line 1205c. The mixing vessel 1208 mixing fragrance material, polymer and/or wax is heated through heater 1221 which is controlled through control line 1221c. The energy of mixing in mixing vessel 1208 is controlled through control line 1208c. Surfactant contained in container 1209 is fed through line 1214 past valve 1211 into mixing vessel 1215, and water or aqueous solution from container 1210 is heated using heater 1230 and flows past valve 1212 into mixing vessel 1215. The flow of the aqueous solution from container 1210 past valve 1212 is controlled through control line 1212c. The amount of heat energy into the aqueous solution in container 1210 is controlled through control line 1230c. The rate of flow of surfactant from container 1209 into mixing vessel 1215 is controlled through control line 1211c. The heat input into mixing vessel 1215 through heater 1231 is controlled through control line 1231c. The surfactant/aqueous solution mixture created in container 1215 is passed through line 1217 into blender 1220. The mixing energy in blender 1220 is controlled through control line 1220c. The flow of product from container 1208 into blending vessel 1220 (e.g., homogenizer) through line 1219 past valve 1218 is controlled through control line 1218c The control of surfactant/aqueous composition from mixing vessel 1215 through line 1217 past valve 1216 into blender 1220 is controlled through control line 1216c. The flow of microemulsion from blender 1220 through line 1223 past control valve 1222 is controlled through control line 1222c, and the cooling energy using cooler 1224 for solid phase particle formation vessel 1225 is controlled through control line 1224c. The mixing energy in the solid phase particle formation vessel 1225 is controlled through control line 1225c. The flow from the solid phase particle formation vessel 1225 to utilization/storage/ inventory vessel 1228 is controlled through control line 1226c, and marketing input information and output information are gathered through control line 1228c.

[0163] By the same token, the apparatus of Figure 12B can be used in conjunction with electronic program controller 1302 which uses marketing input information from source 1301 via a control line feeding into the electronic program controller 1302 and controlling the apparatus as illustrated in Figure 12B and Figure 12D via control lines. Thus, the apparatus shown in schematic diagram in Figure 12B is also shown in schematic form in Figure 12D as associated with the electronic program controller (computer mechanism) via control lines.

[0164] More specifically, fragrances material from container 1250 passing through line 1259 past valve 1258 has its flow controlled through control line 1258c. The polymer and/or wax from container 1251 passing through line 1256 past control valve 1257 has its flow controlled through control line 1257c. Simultaneously, the heat input into container 1251 for heating the polymer and/or wax material using heater 1252 is controlled through control line 1252c. Simultaneously, surfactant from container 1253 flowing through line 1254 past valve 1255 has its flow controlled through control line 1255c. The surfactant, the polymer and/or wax and the fragrance material are mixed in mixing vessel 1260, and the mixing energy is controlled through control line 1260c and the heat input to the mixing vessel is controlled through control line 1261c. Aqueous composition is heated in container 1264 through heating element 1265 which is controlled through control line 1265c. The aqueous composition flow through line 1266 past valve 1267 is controlled through control line 1267c. The mixture of fragrance material, polymer and/or wax and surfactant in mixing vessel 1260 then flows through line 1262 past control valve 1263, and the flow of this mixture into blender (e.g., homogenizer) 1268 is controlled through control line 1263c. The flow of aqueous composition into the blender (e.g., homogenizer) from container 1264 through line 1266 past valve 1267 is controlled through control line 1267c. The homogenizing energy for blender 1268 is controlled through control line 1268c. The product emanating from blender 1268 through line 1269 past valve 1270 has its flow controlled through control line 1270c. The solid phase particle formation component of the apparatus of our invention 1271 is equipped with cooling coils 1272, controlled through control line 1272c. Operation of the solid phase particle formation component of the apparatus of our invention is controlled through control line 1271c. Flow from the solid phase particle formation vessel into the utilization of slurry, storage, inventory and marketing vessel 1275 past valve 1273 through line 1274 is controlled through control line 1273c. Marketing input and output to location 1275 is controlled through control line 1275c.

[0165] Referring to Figure 13, the wax phase from location 1306 is admixed with fragrance oil and optional additives at location 1308 and fed into vessel 1309, simultaneously with water with emulsifier heated to 90°C from location 1305 mixed with surfactant from location 1307 also flowing into vessel 1309 Fluid from vessel 1309 is passed through mixer or homogenizer 1310 and recycled back into vessel 1309 for a given period of time, e.g., one minute. The resulting fluid shown at location 1311 is then again passed through homogenizer 1312 and then quenched to solidify microparticles using a heat exchanger at location 1313.

[0166] Referring to Figure 14, reference numeral 1403 refers to the data points for fragrance A-1. Reference numeral 1404 refers to the data points for fragrance A-2. Reference numeral 1405 refers to the data points for fragrance A-3. Reference numeral 1406 refers to the graph for data points 1403, 1404 and 1405. The **Y** axis is referred to using reference numeral 1401 for the $\{\log_{10}[VP]\}$ (of vapor pressure). The **X** axis is indicated by reference numeral 1402 and

is for the accumulated sum (in weight percent) of the fragrance ingredients, fragrances A-1, A-2 and A-3. The graph 1406 is also described using the equation

$$\{\log_{10}[VP]\} = (0 \cdot 046)S^3 + (1 \cdot 673)S^2 - (16 \cdot 41)S - 4$$

wherein S represents the accumulated sum (weight percent) of the fragrance ingredients and $\{\log_{10}[VP]\}$ represents the log of the vapor pressure of each of the fragrance ingredients.

[0167] Referring to Figure 15, the **Y** axis is indicated by reference numeral 1501 and represents weight loss (weight percent), normalized and shown by the symbol **W**. The **X** axis represents time and is indicated by reference numeral 1502 with time being shown by the symbol θ. Reference numeral 1503 indicates data points for the 50:50 mixture of high and low vapor pressure substances. Reference numeral 1504 represents data points for low vapor pressure substances (that is, <0.01 mm/Hg); and reference numeral 1505 represents data points for high vapor pressure substances (>0.1 mm/Hg). Reference numeral 1508 is the graph for weight loss versus time for the mixture of high and low vapor pressure substances (50:50) and is represented by the equation:

$$W = 94 \cdot 583 - 0 \cdot 29497\ \theta .$$

[0168] Reference numeral 1509 is the equation for weight loss versus time for low vapor pressure materials and is represented by the equation:

$$W = 98 \cdot 679 - 0 \cdot 09478\ \theta ;$$

and reference numeral 1507 is the graph for weight loss versus time for high vapor pressure materials and is represented by the equation:

$$W = 95 \cdot 679 - 0 \cdot 43843\ \theta .$$

[0169] With respect to Figure 16 concerning the fragrance diffusion evaluation system, the **Y** axis concerning weight loss rate (weight percent per minute) is indicated by reference numeral 1601 and is shown by the term

$$\left(\frac{dW}{d\theta}\right) .$$

The **X** axis representing weight percent in the formula of fragrance A-3 is indicated by reference numeral 1602 and is also represented by the term **W**. Reference numeral 1603 represents the data points for the weight loss rate versus weight percent in the formula as measured by thermal gravimetric analysis (**TGA**). Reference numeral 1604 represents the data points for the weight loss rate versus weight percent in the formula as measured by the fragrance diffusion evaluation system **(FES)** of Figures 10A and 10B. Reference numeral 1604a shows the standard deviation for the data points 1604. Reference numeral 1605 is the graph for weight loss rate versus weight percent in the formula as measured by the fragrance diffusion evaluation system and is represented by the equations:

$$\left(\frac{dW}{d\theta}\right) = 0 \cdot 10675 + 0 \cdot 003965\ W$$

and

$$Ln_e(0 \cdot 10675 + 0 \cdot 00395W) = \theta .$$

[0170] The reference numeral 1606 is for the graph of weight loss rate versus weight percent in the formula as measured by thermal gravimetric analysis and is represented by the equations:

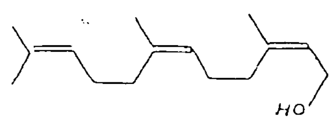
$$\left(\frac{dW}{d\theta}\right) = 0\cdot061929 + 0\cdot00319\ W$$

and

$$Ln_e\ (0{\cdot}061929 + 0{\cdot}00319W) = \theta.$$

[0171] The thermogravimetric analysis (**TGA**) of the prior art is described in detail in Kroschwitz, "Polymers: Polymer Characterization and Analysis" published by John Wiley and Sons, 1990 at pages 837-848 (Chapter entitled: "Thermal Analysis") incorporated by reference herein.

[0172] With reference to Figure 17, the **Y** axis is indicated by reference numeral 1701 and is for aroma chemical loading efficiency indicated by the symbol: ε. The **X** axis is indicated by reference numeral 1702 and is for the $\log_{10}P$ wherein **P** is the n-octanol-water partition coefficient for the materials benzyl alcohol, geraniol, farnesol and GALAXOLIDE®. Reference numeral 1703 is the data point for benzyl alcohol. Reference numeral 1704 is the data point for geraniol. Reference numeral 1704a shows the standard deviation for the data point for geraniol. Reference numeral 1705 is the data point for farnesol having the structure:



Reference numeral 1705a represents the standard deviation for the data point for farnesol. Reference numeral 1706 is the data point for GALAXOLIDE®. Reference numeral 1708 is the graph of aroma chemical loading efficiency versus $\log_{10}P$ wherein **P** is the n-octanol-water partition coefficient for the aroma chemicals The graph is also represented by the equation:

$$\varepsilon = 16{\cdot}1433\ \{\log_{10}P\} - 5{\cdot}922\ .$$

[0173] Aroma chemical loading efficiency is shown by the equation:

$$\varepsilon = \left(\frac{m_C}{m_T}\right) \times 100,$$

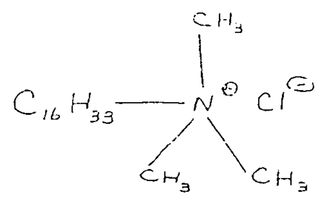
wherein the term: m_C represents the mass of aroma chemicals encapsulated in the microparticles and the term: m_T represents the total mass of the aroma chemical in the microparticle slurry.

[0174] The n-octanol/water partitioning coefficient of a perfume material indicated by the term "**P**" is the ratio between its equilibrium concentrations in n-octanol and in water. The perfume materials of our invention have an n-octanol/water partitioning coefficient **P** of between about 10 and about 10^8. Since the partitioning coefficients of the perfume compositions of this invention have values between 10 and 10^8, they are more conveniently given in the form of their logarithm to the base 10, $\log_{10}P$. Thus, the perfume materials useful in the practice of our invention have a $\log_{10}P$ of between about 1 and about 8 as indicated, supra.

[0175] The $\log_{10}P$ of many perfume ingredients have been reported; for example, the Pomona 92 database, available from Daylight Chemical Information Systems, Inc. (Daylight CIS), Irvine, California, contains many, along with citations to the original literature. However, the $\log_{10}P$ values are most conveniently calculated by the "**CLOGP**" program, also available from Daylight CIS. This program also lists experimental $\log_{10}P$ values when they are available in the Pomona

92 database. The "calculated $\log_{10}P$" is determined by the fragment approach of Hansch and Leo *(Comprehensive Medicinal Chemistry,* Volume 4, C. Hansch, P.G. Sammens, J.B. Taylor and C.A. Ramsden, Editors, page 295, Pergamon Press, 1990, incorporated by reference herein). The fragment approach is based on the the chemical structure of each component of the perfume material and takes into account the numbers and types of atoms, the atom connectivity and chemical bonding. The calculated $\log_{10}P$ values, which are the most reliable and widely used estimates for this physicochemical property, are preferably used instead of the experimental $\log_{10}P$ values in the selection of perfume materials useful in the practice of our invention.

[0176] Figure 18A shows the rotor/stator mixer particle size distribution using 1.2% cetyl trimethyl ammonium chloride having the structure:

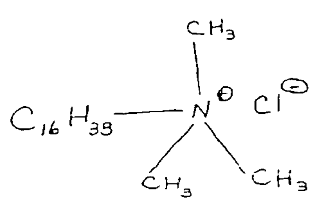


,

10% candelilla wax and 10% fragrance P-50448. The particle size distribution is set forth using reference numeral 1803. The **X** axis is represented by reference numeral 1802 which sets forth the particle diameter in microns. The **Y** axis sets forth the volume percent for each particular particle diameter and is represented by reference numeral 1801. The mean particle size of the particles is 2.4 microns and the distribution is as follows:

90% of particles are finer than:	3.8 microns;
75% of particles are finer than:	2.8 microns,
50% of particles are finer than:	2.0 microns,
25% of particles are finer than:	1.4 microns; and
10% of particles are finer than :	1.1 microns.

[0177] Referring to Figure 18B, Figure 18B shows a homogenized particle size distribution using a mixture containing 0.5% cetyl trimethyl ammonium chloride having the structure:



,

10% candelilla wax and 10% fragrance IB-X-016. The particle size distribution is as follows:

mean particle size: 0.74 microns;

90% of particles are finer than:	2.60 microns;
75% of particles are finer than:	0.70 microns;
70% of particles are finer than:	0.19 microns;
25% of particles are finer than:	0.14 microns, and
10% of particles are finer than:	0.12 microns.

The **X** axis is represented by reference numeral 1812 and indicates particle diameter in microns. The **Y** axis is indicated by reference numeral 1811 and indicates volume percent of particles of particular particle diameter. Reference numeral 1813 shows those particles having a particle diameter of from zero up to about 0.4 microns. Reference numeral 1814

shows those particles having a particle diameter of from about 0.4 up to about 1 micron. Reference numeral 1815 shows those particles having a particle diameter of from about 1.3 up to about 1.6 microns.

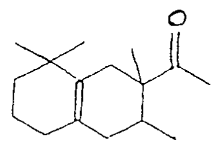
[0178] Referring to Figure 19, the **Y** axis is indicated by reference numeral 1906 and the **X** axis indicating time in minutes is indicated by reference numeral 1905. Reference numeral 1901 represents data points for air flow in the second of two cylinders. Reference numeral 1902 indicates data points for air flow in the first of the two cylinders of Figure 10B. Reference numeral 1903 shows the graph of time versus air flow for the first of the two cylinders in Figure 10B. Reference numeral 1904 sets forth the graph of air flow versus time in the first of the two cylinders as set forth in Figure 10B.

[0179] The following examples illustrate embodiments of this invention. All parts, percentages and proportions referred to herein and in the appended claims are by weight, unless otherwise indicated.

EXAMPLE A

FRAGRANCE COMPOSITION

[0180] The following fragrance composition is prepared for use in Examples I-IV, infra.

Ingredients	Parts by Weight
hexyl cinnamic aldehyde	15
cis-3-hexenyl salicylate	9
ISO E SUPER® (Trademark of International Flavors & Fragrances Inc. of New York, NY) having the structure: 	9
LILIAL® (Trademark of Givaudan, Inc. of Clifton, NJ)	30
β-ionone	8
γ-methyl ionone	6
citronellol	25
methyl nonyl acetaldehyde	15
allyl cyclohexyl propane	5
α-terpineol	6
borneol	5
β-phenyl ethyl alcohol	25
linalool	9
allyl amyl glycolate	7
linalyl acetate	12
dihydromyrcenol	5
isobornyl acetate	20
methyl chavicol	5
benzyl acetate	9
camphor	15
styralyl acetate	5
eucalyptus oil	13
cis-3-hexenyl acetate	4

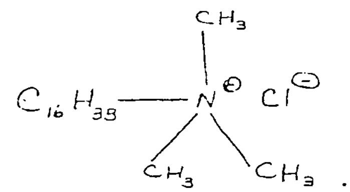
EXAMPLE I

PREPARATION OF MICROPARTICLES USING SILVERSON L4R LABORATORY MIXER OF FIGURE 11G

[0181] The following procedure is used for the preparation of microparticles with the fragrance of Example A and candelilla wax using a Silverson L4R laboratory mixer as set forth in Figure 11G and described, supra. The resulting

formulation is:

84.7% water;
10% candelilla wax;
5% fragrance of Example A; and
0.3% cetyl trimethyl ammonium chloride having the structure:



(1) 37.5 Grams of candelilla wax is placed in an oven at 125°C and allowed to melt.

(2) 314.87 Grams of deionized water is placed into a steam jacket in a one gallon tank.

(3) The bottom of the tank is piped into the suction side of a Silverson in-line model L4R laboratory rotar/stator mixer. The discharge of the mixer was piped back into the tank to allow for recirculation.

(4) The mixer is turned on slowly and the water is drawn into the mixer and pumped back into the tank.

(5) 3.88 Grams of 29% cetyl trimethyl ammonium chloride solution in water is added to the water.

(6) Steam is turned on the jacket and the water/surfactant solution is heated to 90°C. A counter-rotating propeller mixer mounted in the tank ensures that the temperature of the water is homogeneous.

(7) Candelilla wax is removed from the oven and 18.75 grams of the fragrance of Example A is mixed into the wax by hand with a glass rod.

(8) The fragrance/wax mixture is poured into the tank. The counter-rotating propeller mixer speed is increased to disperse the wax/oil into the water and keep the emulsion homogeneous.

(9) The mixer is turned on maximum speed and is allowed to emulsify for one minute. The steam rate is adjusted to maintain a product temperature of 90°C.

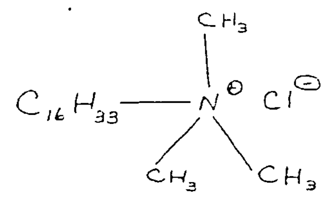
(10) The mixer speed is reduced to a minimum and the three-way valve located on the mixer discharge is turned to divert the emulsion through a Parker dual heat transfer coil to solidify the emulsified wax and reduce the slurry temperature to ambient.

EXAMPLE II

PREPARATION OF MICROPARTICLES USING A GAULIN 15MR HOMOGENIZER AS SET FORTH IN FIGURE 11D AND USING THE HOMOGENIZER PRESSURE ADJUSTMENT VALVE SYSTEM OF FIGURE 11F

[0182] The following procedure is for the preparation of microparticles with the fragrance of Example A in candelilla wax using a Gaulin 15MR homogenizer. The formulation is:

84.7% water;
10% candelilla wax;
5% fragrance of Example A; and
0.3% cetyl trimethyl ammonium chloride having the structure:



The steps are as follows:

(1) 75 Grams of candelilla wax is placed in an oven at 125°C and allowed to melt.

(2) 629.74 Grams of deionized water is placed into a steam-jacketed one gallon tank.

(3) The bottom of the tank is piped into the suction side of a Gaulin 15MR-8TA laboratory homogenizer. The discharge of the homogenizer is piped back into the tank to allow for recirculation.

(4) The homogenizer is turned on with the secondary pressure set at 500 psig. The water is drawn into the homogenizer and pumped back into the tank.

(5) 7.78 Grams of a 29% active cetyl trimethyl ammonium chloride solution in water is added to the water.

(6) Steam is turned on the jacket and the water/surfactant solution is heated to 90°C. A counter-rotating propeller mixer mounted in the tank ensures that the temperature of the water is homogeneous.

(7) The candelilla wax is removed from the oven and 37.5 grams of the fragrance of Example A is mixed with the wax by hand with a glass rod.

(8) The fragrance/wax mixture is poured into the tank. The counter-rotating propeller mixer speed is increased to disperse the wax/oil into the water and keep the emulsion homogeneous.

(9) The second-stage pressure of the homogenizer is set at 6,000 psig and it is allowed to emulsify for one minute Steam rate is adjusted to maintain 90°C product temperature.

(10) A three-way mixing valve located on the homogenizer discharge is turned to divert the emulsion through a Parker dual heat transfer coil to solidify the emulsified wax and to reduce the slurry temperature to ambient.

[0183] The products produced in Examples I-IV, supra, give rise to aesthetically pleasing, long lasting fragrance effects when used in hair care preparation in accordance with the use of Examples:

U.S. Letters Patent No. 5,653,968 issued on August 5, 1997, entitled "RINSE-OFF HAIR CARE COMPOSITIONS"; and

U.S. Letters Patent No. 5,653,969 issued on August 5, 1997, entitled "LOW RESIDUE HAIR CARE COMPOSITIONS".

EXAMPLE III

USE OF SHAMPOO/CONDITIONER

[0184] 0.98 Grams of the slurry of Example I is admixed with 14 grams of a shampoo composition, as described in U.S. Letters Patent No. 5,658,868 issued on August 19, 1997, incorporated by reference herein and containing:

5% (weight) 2-decenyl sulfonate;
15% (weight) sodium sulfosuccinate ester of n-decanolamide;
25% (weight) lauroamphocarboxyglycinate;

4% (weight) coconut amide;
3% (weight) glycol distearate;
4% (weight) aloe vera;
1% (weight) wheat germ oil; and
43% (weight) water.

[0185] The resultant mixture is applied to hair in a washing procedure. The hair is left to dry. After 24 hours, the dried hair thus washed has an aesthetically pleasing aroma having:

(1) a substantivity of 9 on a scale of 1-10;

(2) a quality of 10 on a scale of 1-10; and

(3) an intensity of 3 on a scale of 1-10.

EXAMPLE IV

USE OF FABRIC SOFTENER

[0186] 2.25 Grams of the slurry of Example II is admixed with 25 grams of a fabric softener composition as described in U.S. Letters Patent No. 5,656,585 issued on August 12, 1997 and incorporated by reference herein.

[0187] 100 Grams of unfragranced powder detergent as described in U.S. Letters Patent No. 5,658,875 issued on August 19, 1997, the specification for which is incorporated by reference herein; and the above fabric softener mixture are then placed in a KENMORE® washing machine containing 14 hand towels (fabricated with cotton fabric, having the dimensions: 6" x 6" and weighing 100 grams each).

[0188] After the washing procedure is carried out, the towels are line-dried for a period of 24 hours. 24 Hours thereafter, each of the thus-dried towels has an aesthetically pleasing aroma having:

(1) a substantivity of 9 on a scale of 1-10;

(2) a quality of 10 on a scale of 1-10; and

(3) an intensity of 5 on a scale of 1-10.

[0189] The features disclosed in the foregoing description, in the following claims and/or in the accompanying drawings may both separately and in any combination thereof, be material for realising the invention in diverse forms thereof.

Claims

1. A composition for fragrancing at least one perfumable material and the environment proximate thereto comprising at least one substantially ellipsoidal hydrophobic particle having a continuous outer surface and an internal matrix volume consisting essentially of:

   (i) a single phase solid solution of a matrix material selected from the group consisting of at least one hydrophobic polymer and at least one hydrophobic wax, each of which polymer and wax has a melting point in the range of from about 35°C up to about 120°C at 1 atmosphere pressure, having dissolved therein at least one hydrophobic fragrance material, said solid solution having an outer surface and an internal matrix volume; and

   (ii) proximate to substantially the entirety of said outer surface, a substantially hydrophilic surfactant,

   said fragrance material having a calculated $\mathbf{log_{10}P}$ in the range of from about 1 up to about 8 wherein **P** is the partition coefficient of the fragrance material between n-octanol and water, said hdyrophobic particle having an outside diameter in the range of from about 0.05 up to about 20 microns, the concentration of fragrance material in said polymer or said wax being from about 5% up to about 60% by weight of said particle, the weight percent of said surfactant being from about 0.01% up to about 5% by weight of said particle, with said wax, said surfactant and said polymer each being nonreactive with said fragrance material and one another.

2. The composition of Claim 1 wherein the permeation rate of said fragrance material through said wax or said polymer is in the range of from about $10^{-8} \frac{mg \cdot mm}{cm^2 \cdot min}$ up to about $8 \times 10^{-3} \frac{mg \cdot mm}{cm^2 \cdot min}$ as determined by the IFF permeation test.

3. The composition of Claim 1 wherein the substantially hydrophilic surfactant is substantially entirely coated on and fixedly bonded to the entirety of said outer surface of said single phase solid solution in the form of a continuous submicron layer of surfactant.

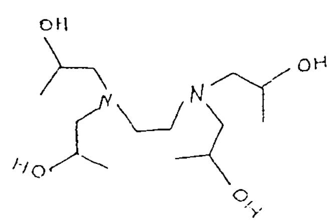
4. The composition of Claim 1 wherein the substantially hydrophilic surfactant is located proximate to and immediately substantially beneath the entirety of said outer surface of said solid solution; and substantially within said internal matrix volume.

5. The composition of Claim 1 wherein the substantially hydrophilic surfactant is both (a) substantially entirely coated on and fixedly bonded to the entirety of said outer surface of said single phase solid solution in the form of a continuous submicron layer of surfactant, and (b) located proximate to and immediately, substantially beneath the entirety of said outer surface of said solid solution and substantially within said internal matrix volume.

6. The composition of Claim 1 wherein the surfactant is selected from the group consisting of:

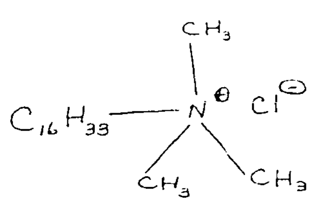
(a) the cationic modified starch, RediBOND® 5320 (trademark of the National Starch Company of Bridgewater, New Jersey), in admixture with partially hydrolyzed polyvinyl acetate, being hydrolyzed from about 73% up to about 99% and having a molecular weight in the range of from about 5,000 up to about 67,000,

(b) the substance tetra(2-hydroxypropyl) ethylenediamine, having the structure



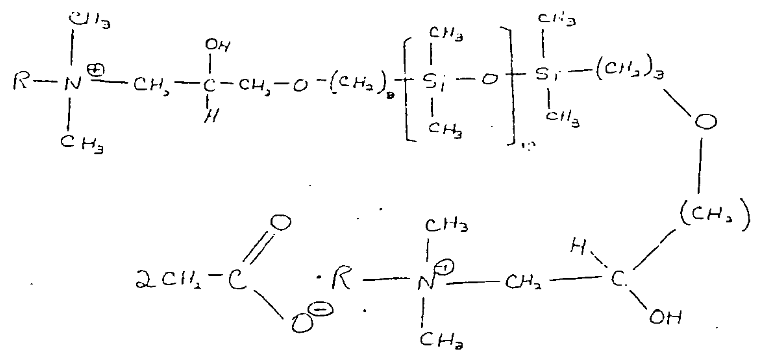
;

(c) cetyl trimethyl ammonium chloride having the structure



;

(d) a quaternary ammonium polysilane derivative having the structure:



wherein **R** is the moiety having the structure:

$$CH_3\text{-}[CH_2]_x\text{-};$$

wherein **m** is an integer of from 10 up to 100; and wherein **x** is an integer of from 10 up to 100, in admixture with partially hydrolyzed polyvinyl acetate, being hydrolyzed from about 73% up to about 99% and having a molecular weight in the range of from about 5,000 up to about 67,000;

and the matrix is selected from the group consisting of:

(a) polyamides having a molecular weight in the range of from about 6, 000 up to about 12,000;

(b) carnauba wax;

(c) candelilla wax;

(d) mixtures of cetyl palmitate and carnauba wax;

(e) mixtures of cetyl palmitate and candelilla wax;

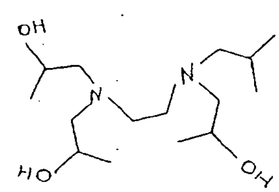
(f) ozokerite wax;

(g) ceresin wax; and

(h) low density polyethylene wax having a molecular weight in the range of from about 500 up to about 6,000.

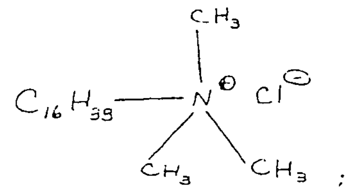
7. The composition of Claim 6 wherein the surfactant is, in the alternative:

(i) tetra(2-hydroxypropyl) ethylenediamine, having the structure:



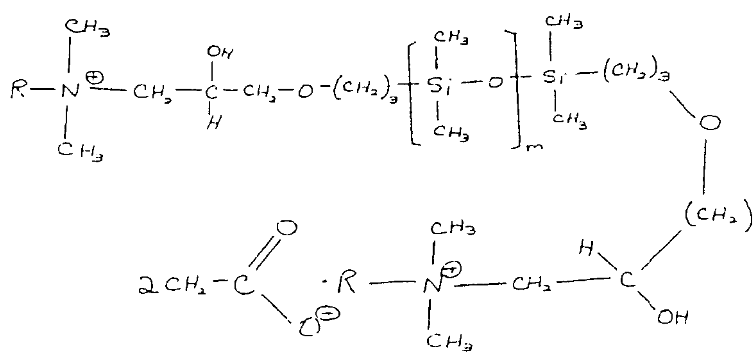
;

(ii) cetyl trimethyl ammonium chloride having the structure.



or

(iii) the quaternary ammonium polysilane derivative having the structure:



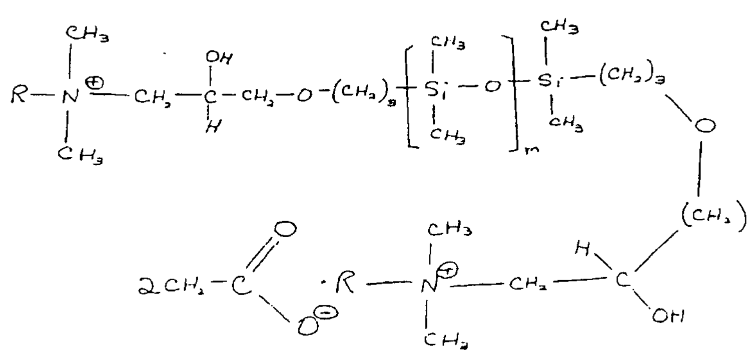
wherein **R** is the moiety having the structure:

$$CH_3\text{-}[CH_2]_x\text{-} ;$$

wherein **m** is an integer of from 10 up to 100; and wherein **x** is an integer of from 10 up to 100, and the weight ratio of quaternary ammonium polysilane derivative:partially hydrolyzed polyvinyl acetate is in the range of from about 2:1 up to about 1:2.

8. A mixture of cationic modified starch, RediBOND® 5320 (trademark of National Starch Inc. of Bridgewater, New Jersey) and partially hydrolyzed polyvinyl acetate, being partially hydrolyzed to an extent of from about 73% up to about 99%, having a molecular weight in the range of from about 5,000 up to about 67,000, with the weight ratio of cationic modified starch:partially hydrolyzed polyvinyl acetate being in the range of from about 2:1 up to about 1:2.

9. A mixture of quaternary ammonium polysilane derivative having the structure:

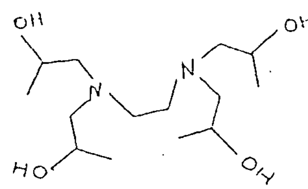


wherein **R** is the moiety having the structure:

$CH_3\text{-}[CH_2]_x\text{-}$;

wherein **m** is an integer of from 10 up to 100; and wherein **x** is an integer of from 10 up to 100, with the weight ratio of quaternary ammonium polysilane derivative:partially hydrolyzed polyvinyl acetate being in the range of from about 1:2 up to about 2:1.

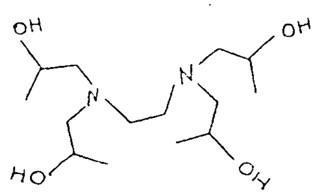
10. A method for increasing the substantivity of a fragrance material selected from the group consisting of at least one aroma chemical and at least one perfume composition comprising the step of intimately admixing with the said fragrance material a substantivity increasing quantity and concentration of tetra(2-hydroxypropyl) ethylenediamine, having the structure:



11. A composition of matter having a high fragrance substantivity comprising in intimate admixture:

(a) a fragrance material selected from the group consisting of at least one aroma chemical and at least one perfume composition; and

(b) tetra(2-hydroxypropyl) ethylenediamine, having the structure:

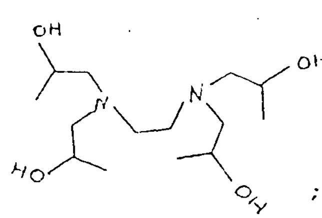


12. A process for fragrancing a perfumable material having a substantially solid surface comprising the step of contacting said solid surface of said perfumable material with at least one particle of Claim 1.

13. The process of Claim 12 wherein the surfactant is, in the alternative:

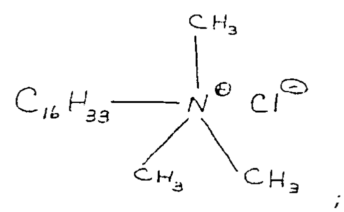
(a) the cationic modified starch, RediBOND® 5320 (trademark of National Starch Inc. of Bridgewater, New Jersey), in admixture with partially hydrolyzed polyvinyl acetate being hydrolyzed to the extent of from about 73% up to about 99%, having a molecular weight in the range of from about 5,000 up to about 67,000;

(b) tetra(2-hydroxypropyl) ethylenediamine, having the structure:



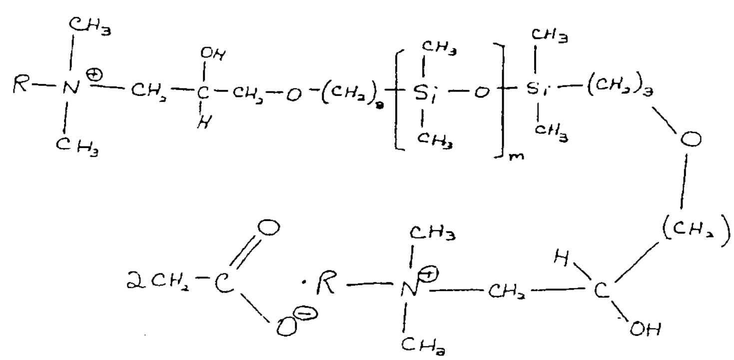
;

(c) cetyl trimethyl ammonium chloride having the structure:



and

(d) a quaternary ammonium polysilane derivative having the structure:



wherein **R** is the moiety having the structure:

$$CH_3\text{-}[CH_2]_x\text{-};$$

wherein **m** is an integer of from 10 up to 100; and wherein **x** is an integer of from 10 up to 100, in admixture with partially hydrolyzed polyvinyl acetate being hydrolyzed to the extent of from about 73% up to about 99%, having a molecular weight in the range of from about 5,000 up to about 67,000,

and the matrix material is selected from the group consisting of:

(a) polyamides having a molecular weight in the range of from about 6,000 up to about 12,000,

(b) carnauba wax;

(c) candelilla wax;

(d) mixtures of cetyl palmitate and carnauba wax;

(e) mixtures of cetyl palmitate and candelilla wax,

(f) ozokerite wax;

(g) ceresin wax; and

(h) low density polyethylene wax having a molecular weight in the range of from about 500 up to about 6, 000.

14. A process for preparing a hydrophobic perfume-containing composition comprising the steps of.

(i) intimately admixing at least one hydrophobic fragrance material with at least one hydrophobic polymer and/

or at least one hydrophobic wax to form a first liquid single phase mixture at a temperature greater than or equal to the melting point of said polymer, said wax or, in the case of mixtures, the melting point of the highest melting point component of the materials in the mixture;

(ii) intimately admixing a surfactant with an aqueous composition comprising water to form a second mixture which is an aqueous solution;

(iii) blending said first mixture and said second mixture at a temperature of between 60°C and the boiling point of said aqueous composition whereby a microemulsion is formed; and

(iv) causing the hydrophobic perfume-containing composition in the solid phase to form as an aqueous suspension of solid phase particles, wherein the weight percent of fragrance for forming said first mixture is in the range of from about 5% up to about 60% by weight of said first mixture; and wherein the weight percent of surfactant in the second mixture is from about 0.01% up to about 5% by weight of said second mixture.

15. A process for preparing a hydrophobic perfume-containing composition comprising the steps of:

(i) intimately admixing at least one hydrophobic fragrance material with (a) at least one hydrophobic polymer and/or at least one hydrophobic wax and (b) at least one surfactant to form a first single liquid phase mixture at a temperature greater than or equal to the melting point of said polymer or said wax or, in the case of mixtures, the melting point of the highest melting point component of the mixture;

(ii) blending said first single liquid phase mixture with an aqueous composition comprising water whereby a microemulsion is formed; and

(iii) causing the hydrophobic perfume-containing composition in the solid phase to form as an aqueous suspension of solid phase particles,

wherein the weight percent of fragrance for forming said first mixture is in the range of from about 5% up to about 60% by weight of said first mixture; wherein the weight percent of surfactant in said first mixture is from about 0.01% up to about 5% by weight of said first mixture.

16. A composition for effecting the targeted delivery of a bioactive or active substance to a substantially solid surface comprising at least one substantially ellipsoidal hydrophobic particle having a continuous outer surface and an internal matrix volume consisting essentially of:

(i) a single phase solid solution of a matrix material selected from the group consisting of at least one hydrophobic polymer and at least one hydrophobic wax, each of which polymer and wax has a melting point in the range of from about 35°C up to about 120°C at 1 atmosphere pressure, having dissolved therein at least one bioactive or active substance, said solid solution having an outer surface and an internal matrix volume; and

(ii) proximate to substantially the entirety of said outer surface, a substantially hydrophilic surfactant,

said bioactive or active substance having a calculated $\mathbf{log_{10}P}$ in the range of from about 1 up to about 8 wherein P is the partition coefficient of the bioactive or active substance between n-octanol and water; said hdyrophobic particle having an outside diameter in the range of from about 0.05 up to about 20 microns; the concentration of active or bioactive substance in said polymer or said wax being from about 5% up to about 60% by weight of said particle; the weight percent of said surfactant being from about 0.01% up to about 5% by weight of said particle; with said wax, said surfactant and said polymer each being nonreactive with said active and said bioactive substances and one another.

17. Apparatus located in an **X-Y-Z** three-space for simultaneously testing the diffusivity, odor character and odor intensity of a fragrance material selected from the group consisting of one or more aroma chemicals and one or more fragrance compositions comprising:

(a) hollow vertically-disposed substantially cylindrical container means having:

(i) a central axis parallel to the **Z** axis;

(ii) an inner void,

(iii) a substantially circular non-interrupted base composed of a first solid nonporous substance located in a first **X-Y** plane, said base being substantially perpendicular to said **Z** axis and having an inner void side and an outside;

(iv) vertically surrounding said inner void and parallel to and substantially equidistant from said **Z** axis, a continuous cylindrical sidewall composed of a second solid nonporous substance having an inner void side and an outside, having a top circular rim located in a second **X-Y** plane, an upper midportion, a lower midportion and a bottom rim located in said first **X-Y** plane, said bottom rim being fixedly sealed along its entire circumference to the circumference of said base, said sidewall having a first orifice therethrough located in the lower midportion thereof in a third **X-Y** plane and a second orifice therethrough substantially diametrically opposed to said first orifice located in a fourth **X-Y** plane, said fourth **X-Y** plane being located between and substantially parallel to said third **X-Y** plane and said first **X-Y** plane, said sidewall otherwise being non-interrupted;

(v) a top substantially circular cover composed of a third solid nonporous substance and having an inner void side and an outside sealably attached along its entire outer circumference to the circumference of the top circular rim of said sidewall and located in said second **X-Y** plane, having a substantially circular top cover opening having an inner circumference, said inner circumference being substantially equidistant from said **Z** axis, said opening having a diameter of from about 20% up to about 40% of the outside diameter of the top cover;

(b) sealably mounted through said first orifice, a temperature probe having a temperature variation-sensitive end attached to support means, said temperature variation-sensitive end being located within said inner void, said support means seabably inserted through said first orifice and connected to temperature monitoring means outside said container means;

(c) sealably mounted through said second orifice, air flow means for supplying a stream of air into said inner void, said air flow means comprising a supply tube having an open end located within said inner void, being connected to said supply tube and located outside said container means. an air supply source; and

(d) suspension means for suspending a test sample located within said inner void, said suspension means comprising a substantially flexible hanging support fixedly attached at two substantially diametrically opposed attachment locations located in substantially the same **X-Y** plane proximate said second **X-Y** plane, each being proximate the top rim of said sidewall and having attached thereto and suspended therefrom at a location substantially midway between said attachment locations, said test sample, initially having absorbed thereon said fragrance material,

whereby when the air supply means is engaged thereby causing air to flow through said air flow means, and the temperature monitor is engaged, air at a fixed temperature **T** or a variable temperature $\mathbf{T(\theta)}$ will flow at a fixed flow rate **Q** or a variable flow rate $\mathbf{Q(\theta)}$ past said sample having an area **A** initially holding $\mathbf{G_o}$ grams of fragrance material (s) in a concentration of $\mathbf{C_o}$ gram-moles per liter for a period of time θ at the end of which time said sample will hold $\mathbf{G_1}$ grams of fragrance material in a concentration $\mathbf{C_1}$ gram-moles per liter during which time θ the odor character and intensity is capable of being measured and determined proximate the intersection of said **Z** axis with said second **X-Y** plane.

18. The apparatus of Claim 17 wherein (i) the temperature of the air supplied by the air supply means is controlled and (ii) the temperature probe has programmed feedback means connected to the air supply means.

19. A process for simultaneously testing the diffusivity, odor character and odor intensity of a fragrance material selected from the group consisting of one or more aroma chemicals and one or more fragrance compositions comprising the steps of:

(a) providing the apparatus of Claim 1;

(b) suspending a test sample containing a fragrance material in the suspension means of said apparatus; and

(c) engaging the air supply means and the temperature monitor means,

whereby when the air supply means is engaged to thereby causing air to flow through the air flow means and the temperature monitor is engaged, air at a fixed temperature **T** or a variable temperature **T(θ)** will flow at a fixed flow rate **Q** or a variable flow rate **Q(θ)** past said sample having an area **A** initially holding G_0 grams of fragrance materials in a concentration of C_0 gram-moles per liter for a period of time θ at the end of which time θ said sample will hold G_1 grams of fragrance material in a concentration C_1 gram-moles per liter during which time θ the odor character and intensity is capable of being measured and determined proximate the intersection of said **Z** axis with said second **X-Y** plane.

20. The apparatus of Claim 17 wherein the container means has:

(i) a height of in the range of from about 50 up to about 75 cm;

(ii) a radius in the range of from about 15 up to about 30 cm;

(iii) a volume in the range of from about 0.1 up to about 0.2 m^3;

(iv) a vertical distance of temperature probe to base of between about 10 and about 30 cm;

(v) a vertical distance of air flow means from base from about 3 up to about 10 cm; and

(vi) a diameter of inner opening in top cover of from about 15 up to about 30 cm.

21. The process of Claim 19 wherein the internal pressure in the inner void is maintained at from about 0.5 up to about 2 psig and the air flow rate is maintained in the range of from about 900 up to about 1,000 ml/minute; and wherein in the apparatus provided, the container means has:

(i) a height of container of from about 50 up to about 75 cm;

(ii) a radius of container means of from about 15 up to about 30 cm;

(iii) a volume of from about 0.1 up to about 0.2 m^3;

(iv) a vertical distance of temperature probe from base of from about 10 and about 30 cm;

(v) a vertical distance of air flow means from base from about 3 up to about 10 cm; and

(vi) a diameter of inner opening in top cover of from about 15 up to about 30 cm.

FIG.1-A

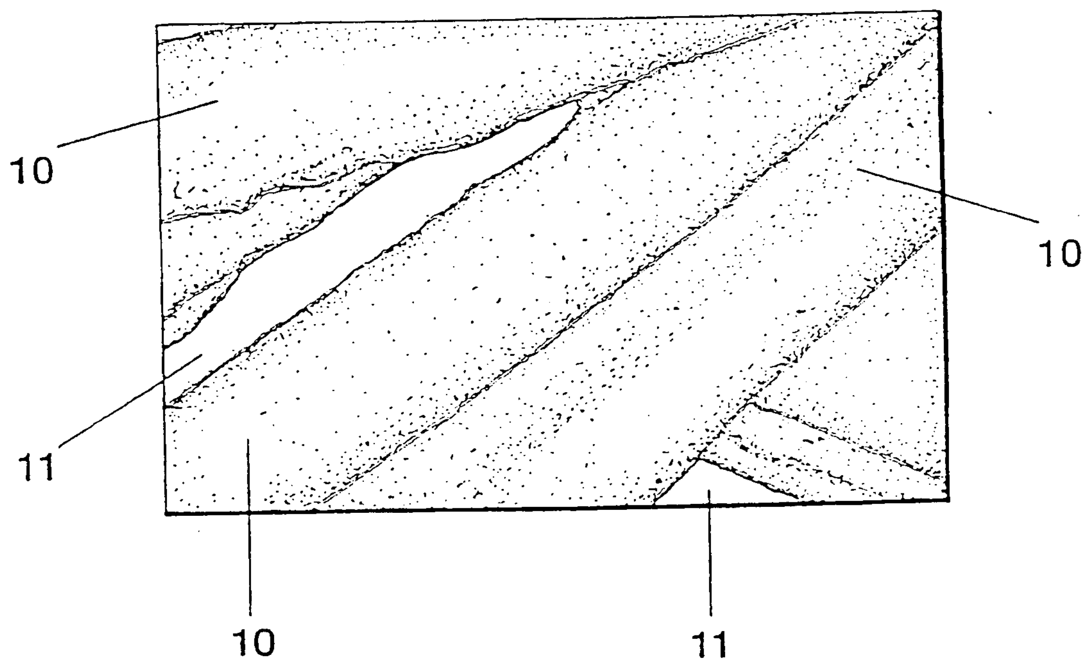


FIG.1-A (I)

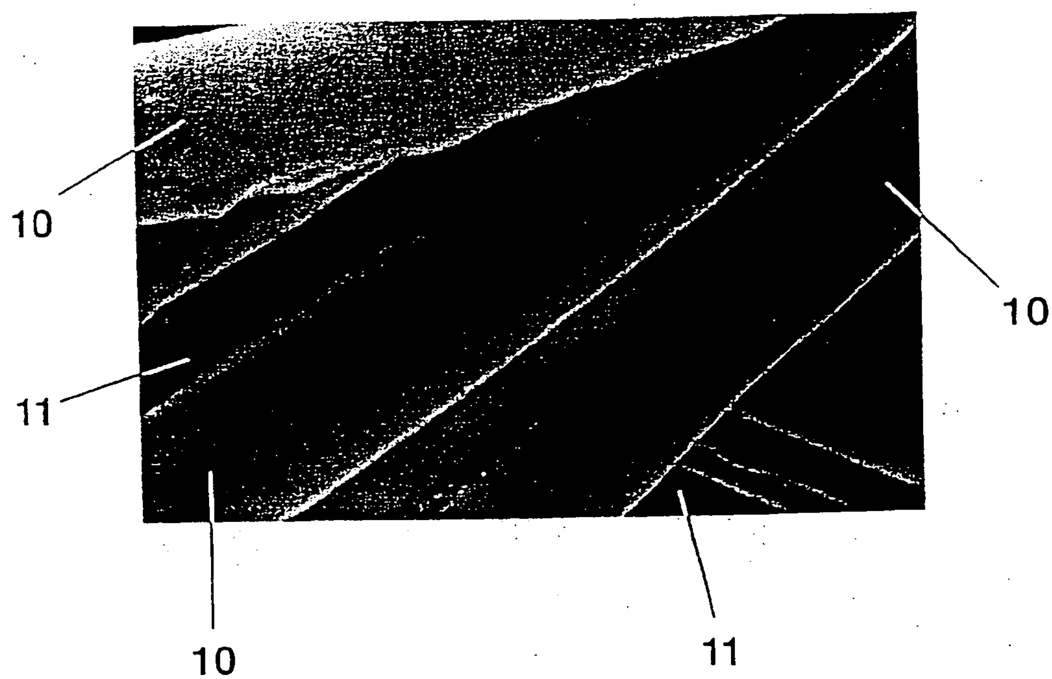


FIG. 1-B

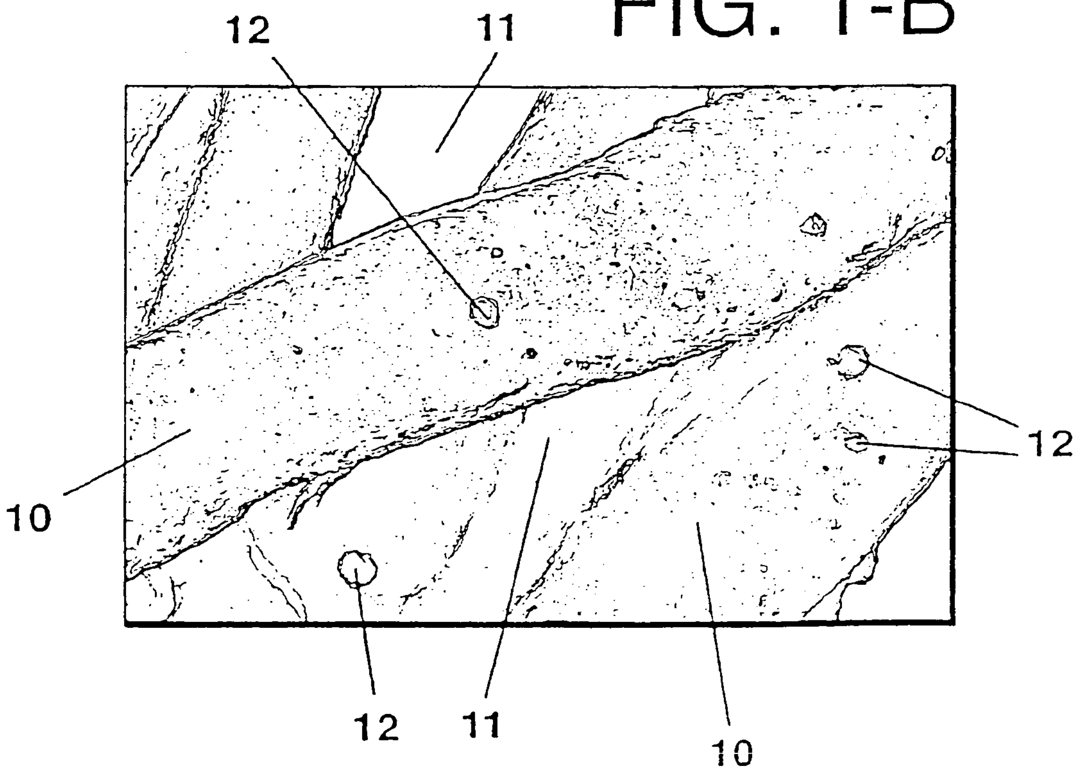


FIG.1-B(I)

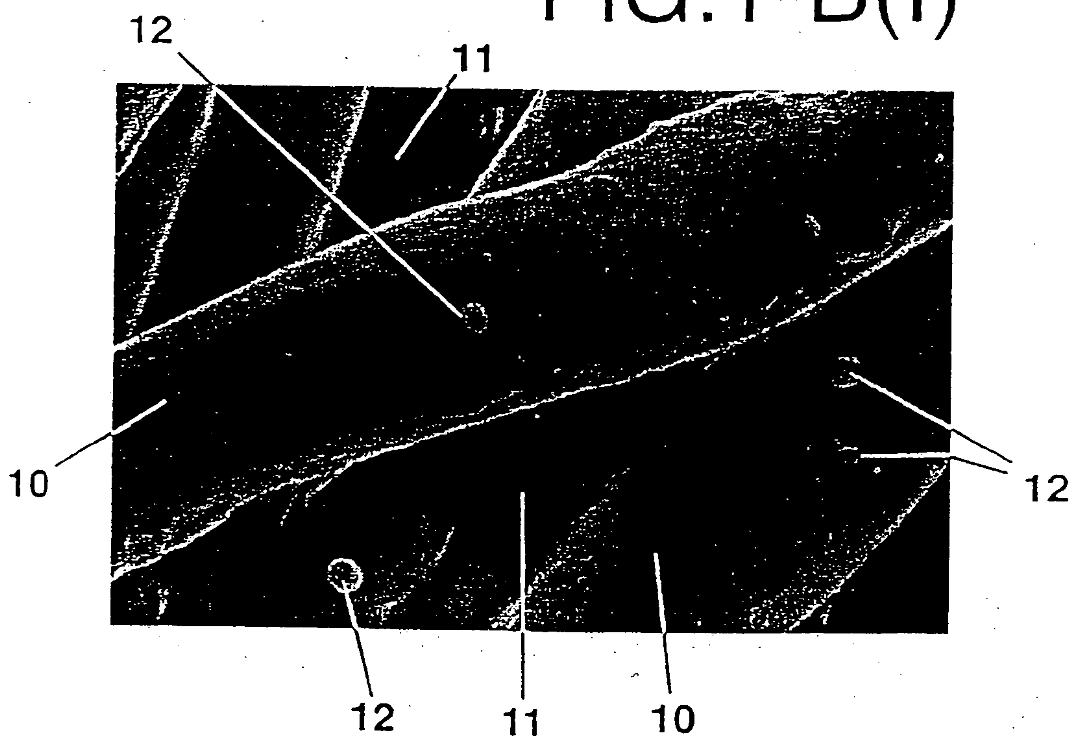


FIG.2-A

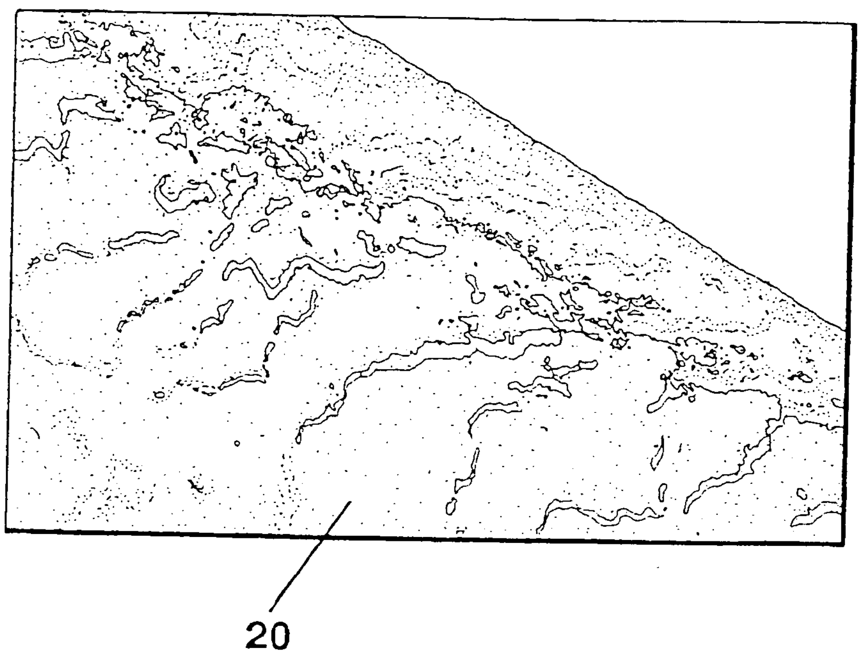
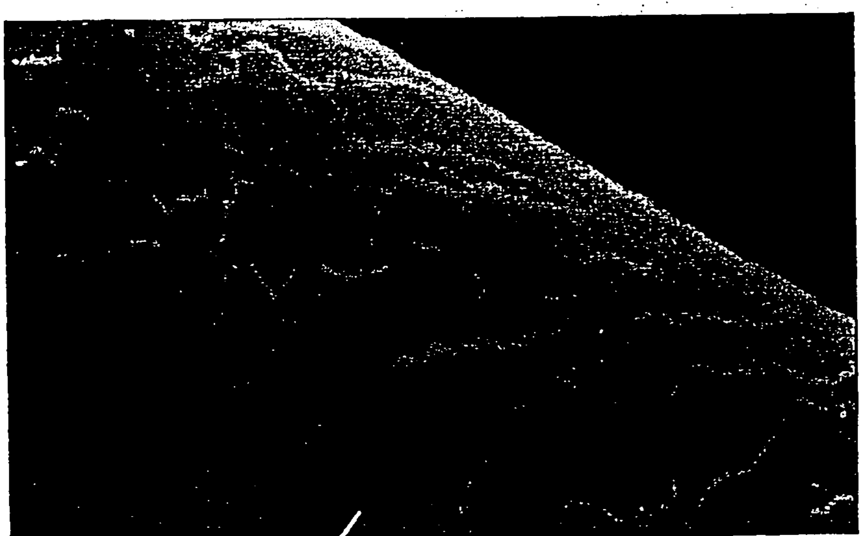


FIG. 2-A(I)



20

FIG.2-B

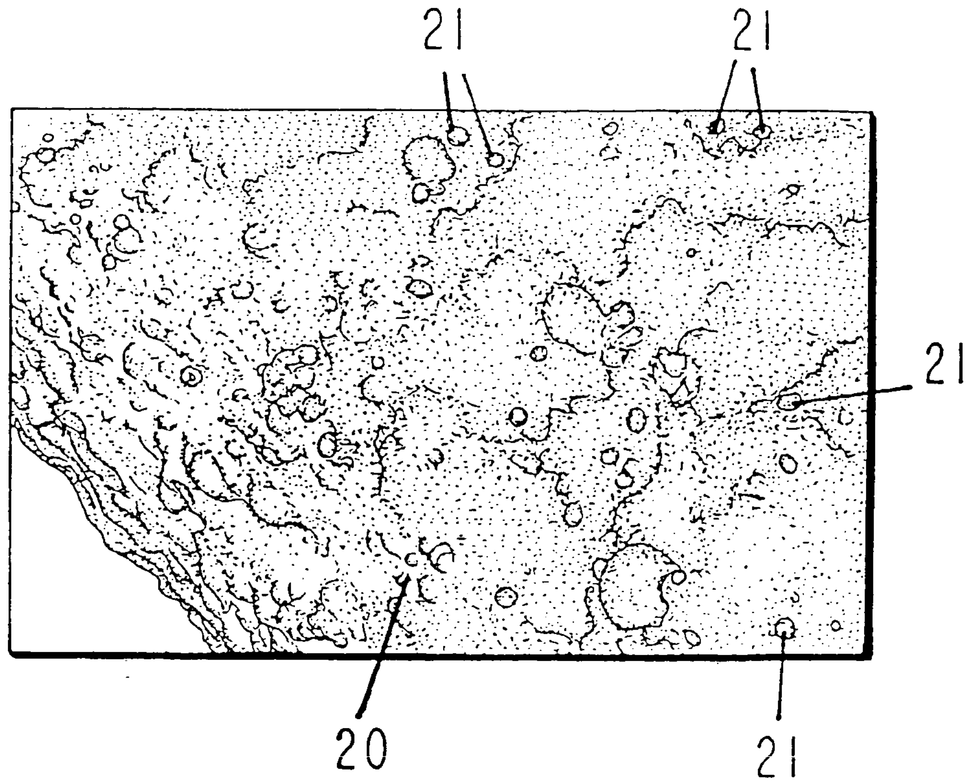


FIG. 2-B(I)

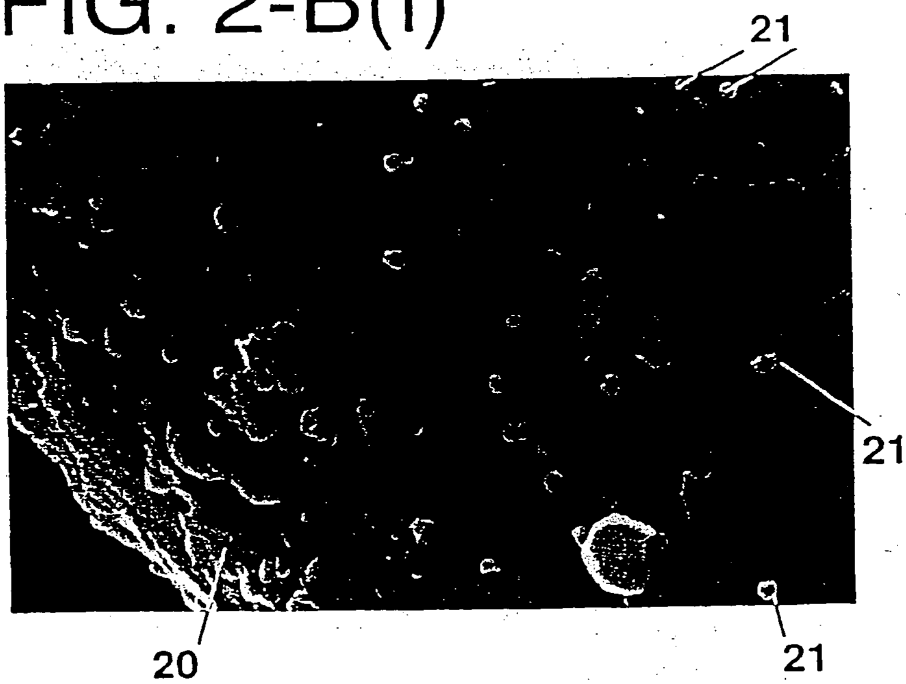


FIG.2-C

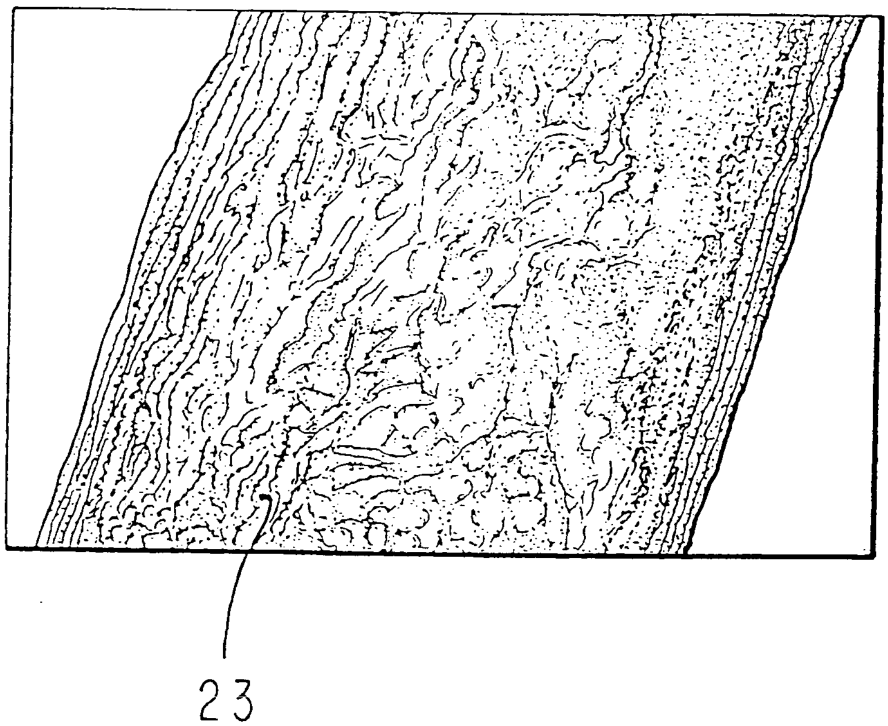
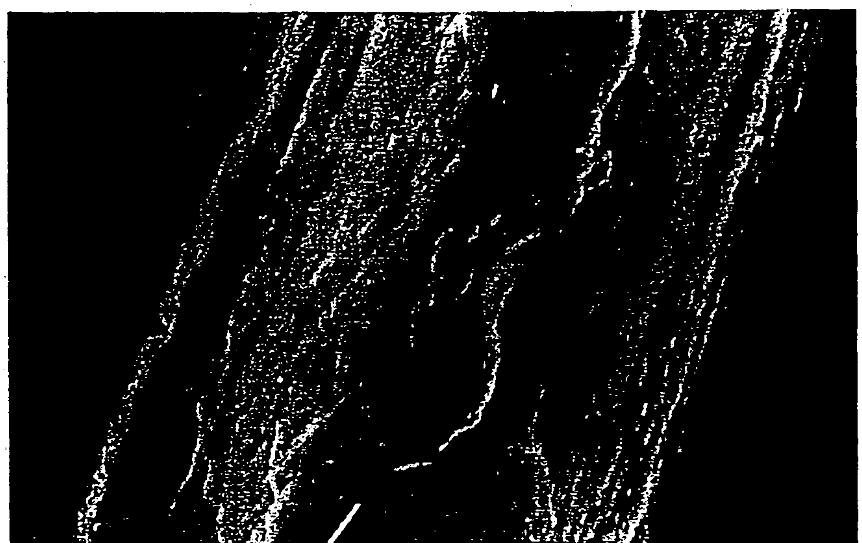


FIG.2-C(I)



23

FIG.2-D

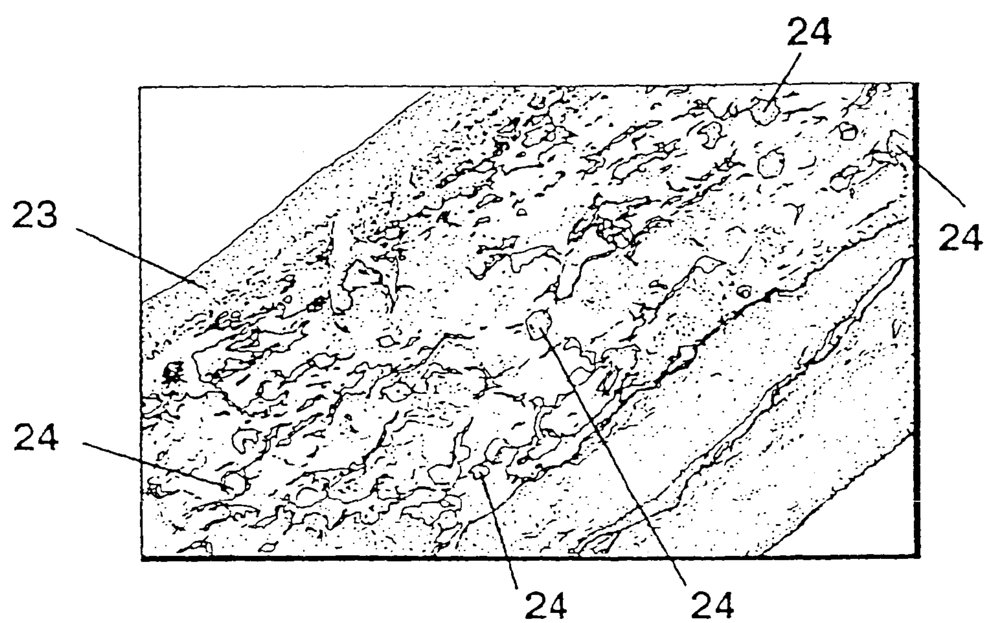


FIG. 2-D (I)

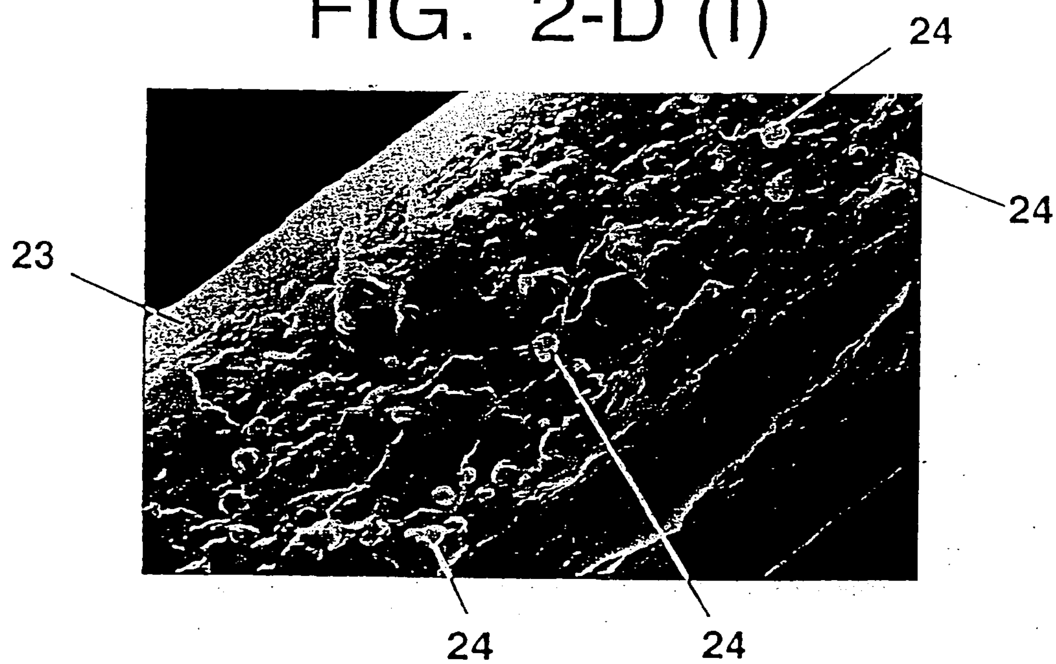


FIG.3-A

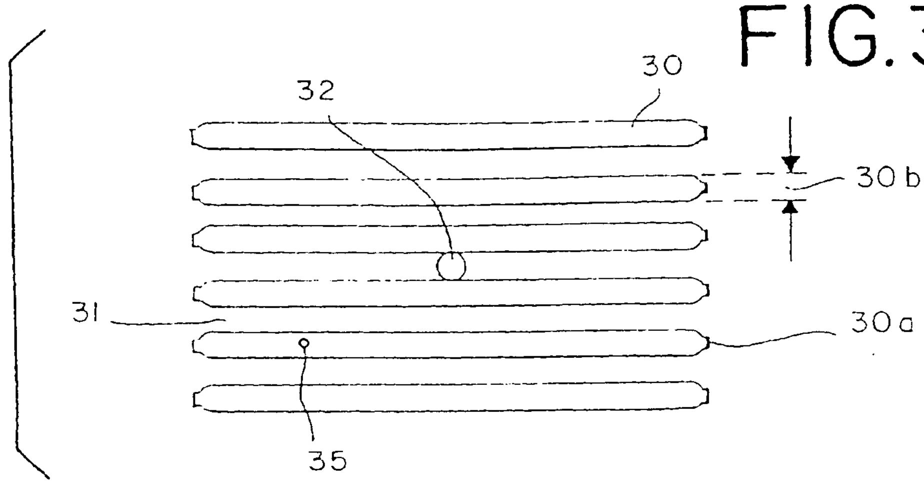


FIG.3-B

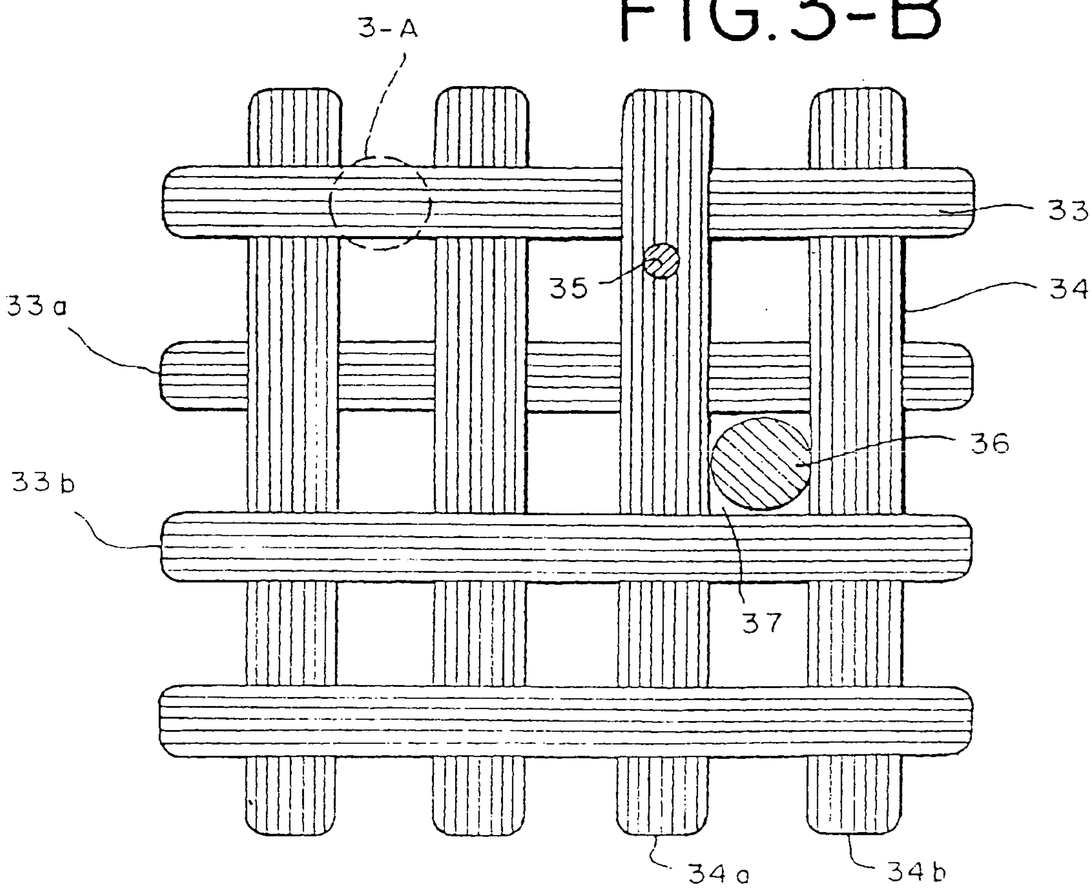


FIG.4-A

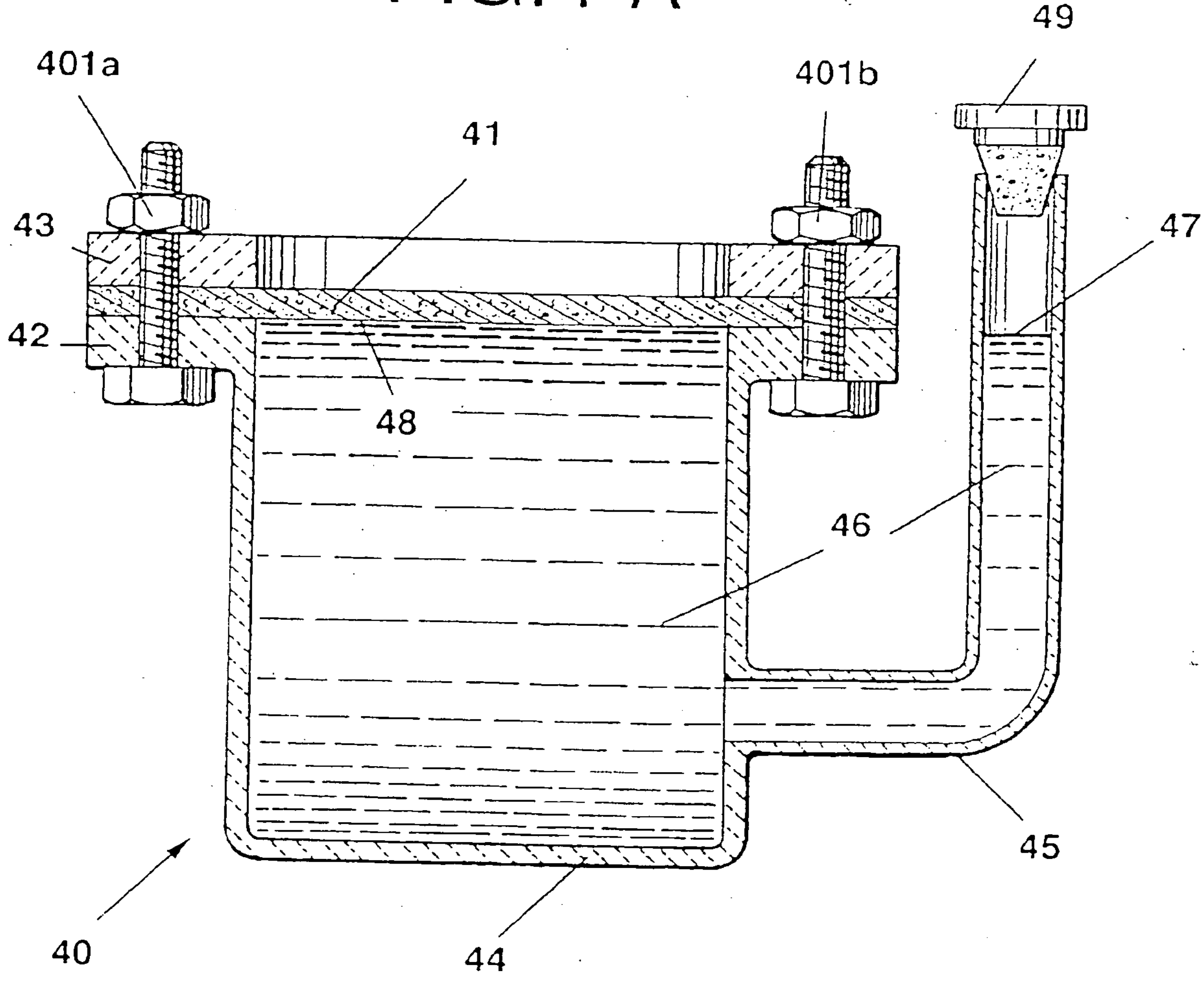


FIG.4-B

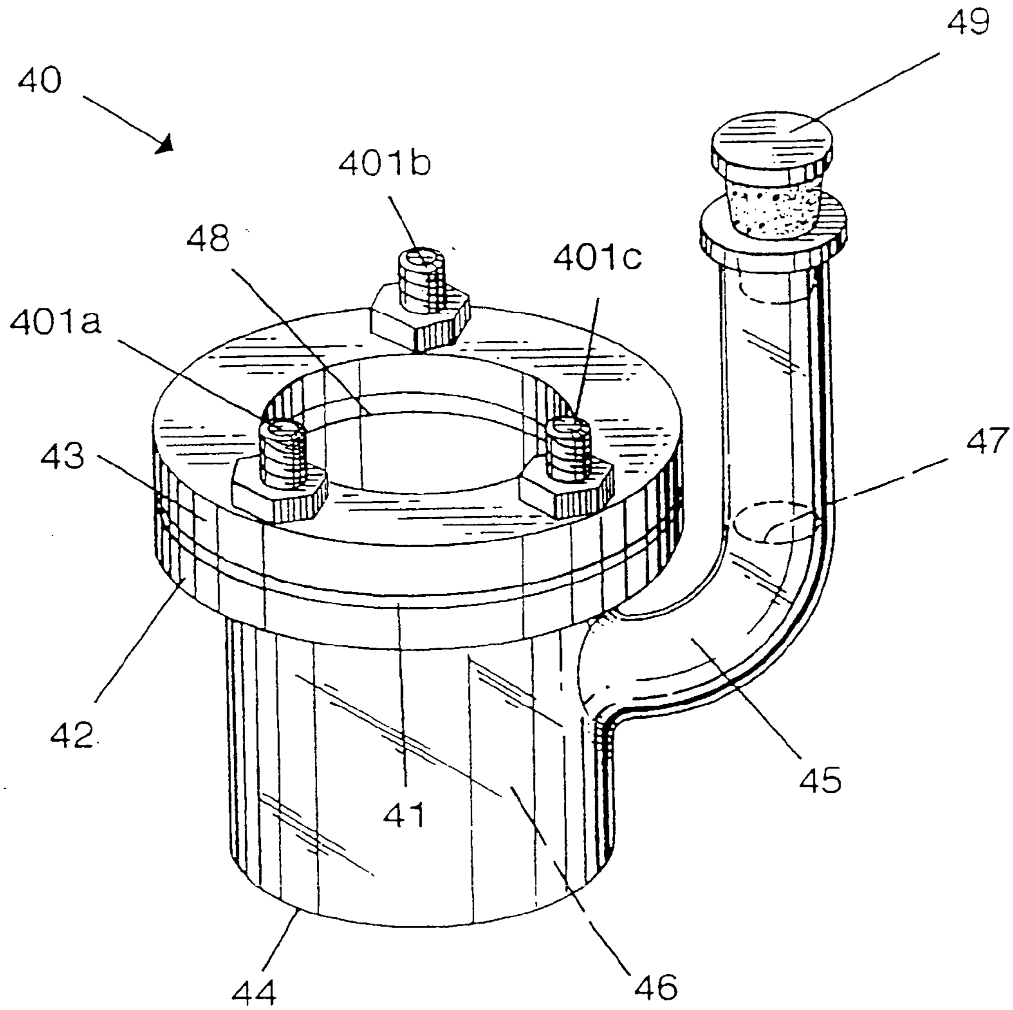


FIG.5-A

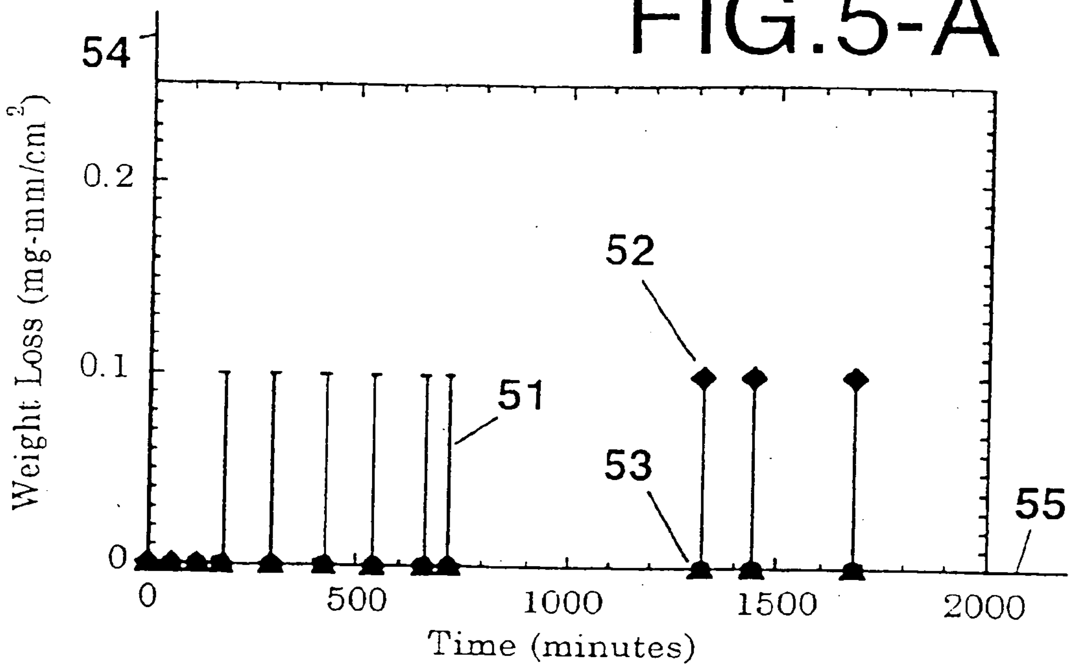

54
Weight Loss (mg-mm/cm^2)
0.2
0.1
0
52
51
53
55
0
500
1000
1500
2000
Time (minutes)

FIG.5-B

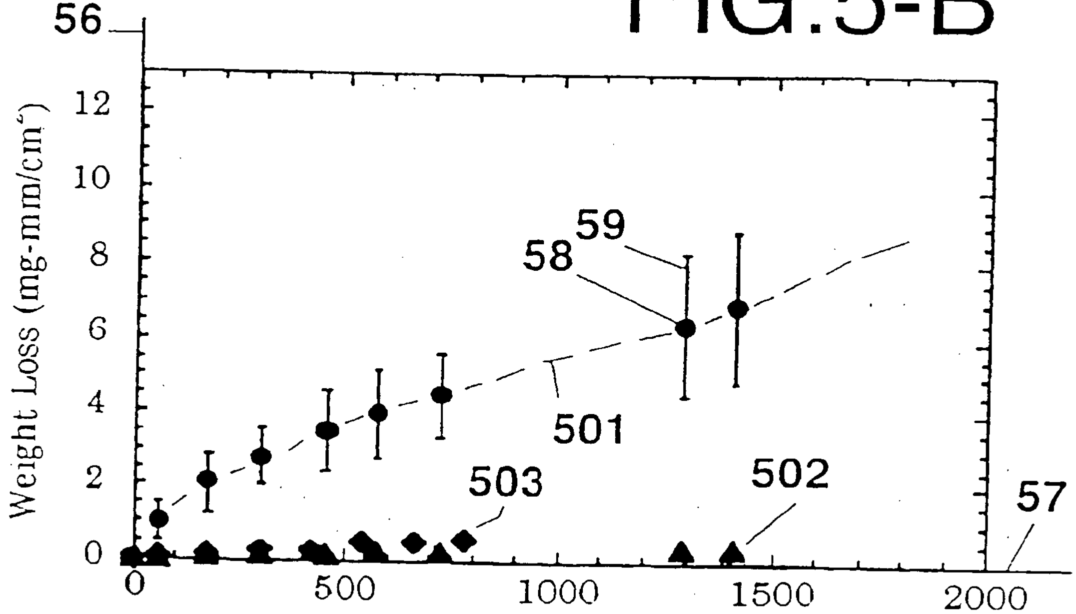

56
Weight Loss (mg-mm/cm^2)
12
10
8
6
4
2
0
59
58
501
503
502
57
0
500
1000
1500
2000

FIG. 5-C

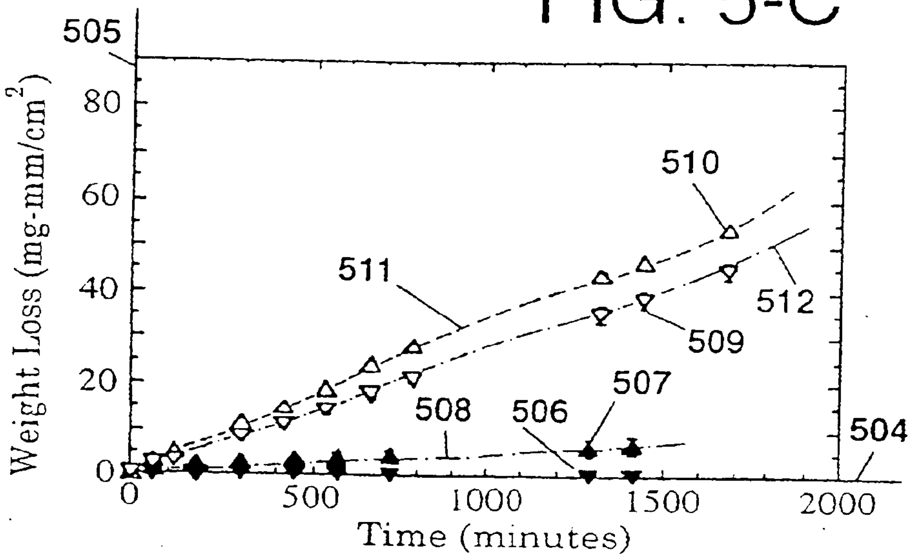
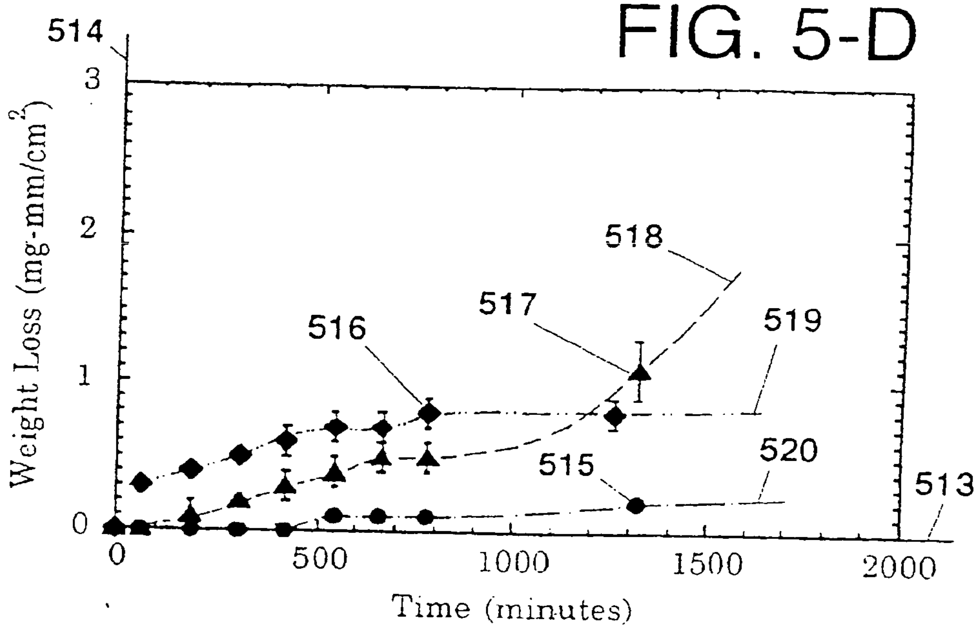

505
80
60
40
20
0
Weight Loss (mg-mm/cm^2)
0
500
1000
1500
2000
Time (minutes)
510
511
512
509
507
508
506
504

FIG. 5-D


514
3
2
1
0
Weight Loss (mg-mm/cm^2)
0
500
1000
1500
2000
Time (minutes)
518
517
516
519
520
515
513

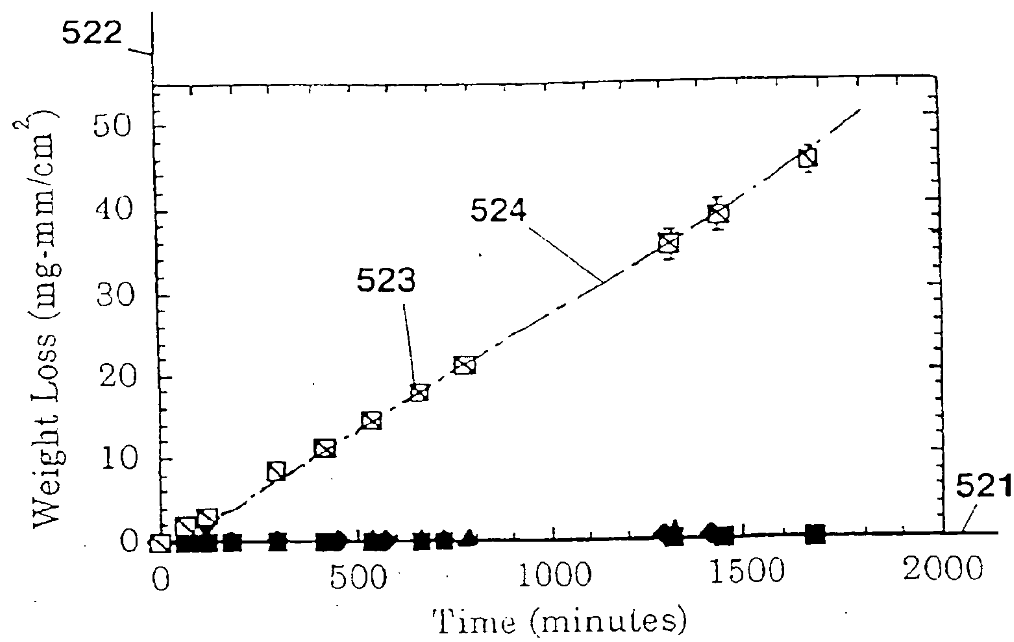


FIG.5-E

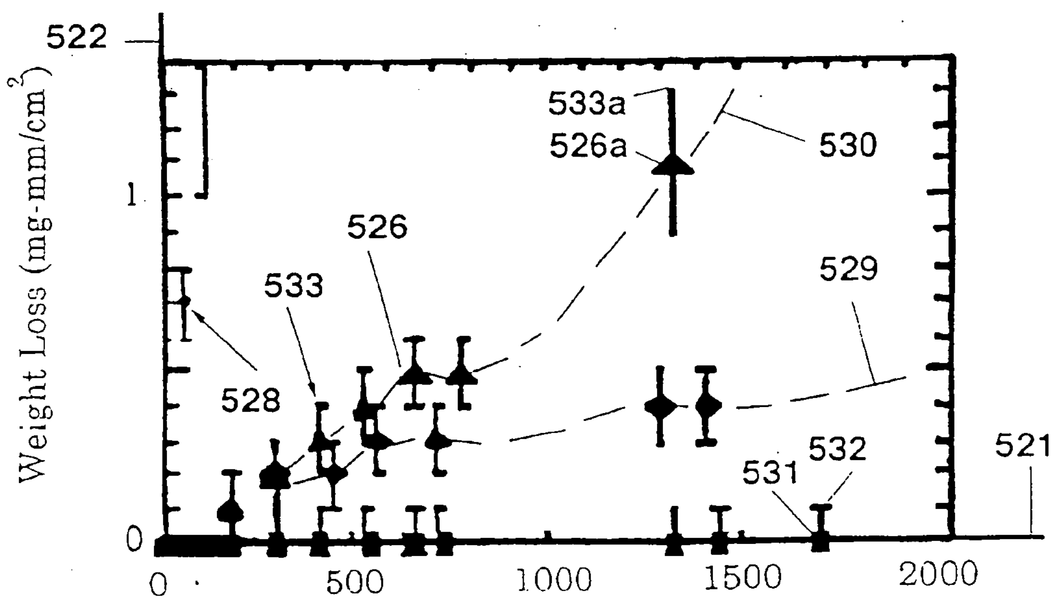


FIG.5-E(A)

FIG. 5-F

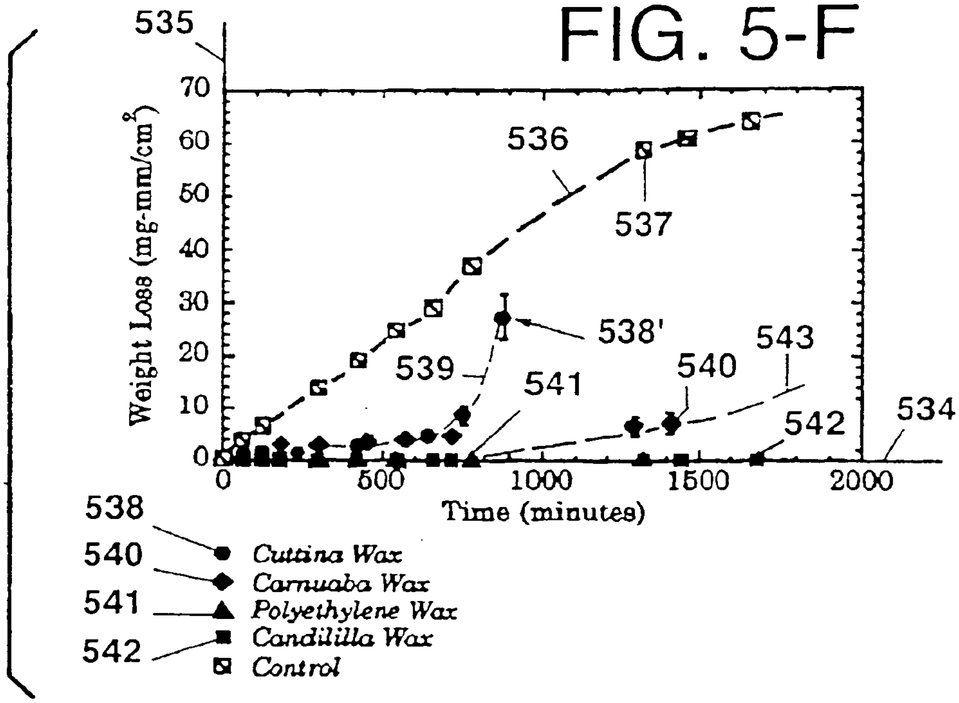

535
70
60
50
40
30
20
10
0
Weight Loss (mg-mm/cm^2)
536
537
538'
539
541
540
543
542
534
0
500
1000
1500
2000
Time (minutes)
538
540
541
542
Cuttina Wax
Carnuaba Wax
Polyethylene Wax
Candilillo Wax
Control

FIG. 5-G

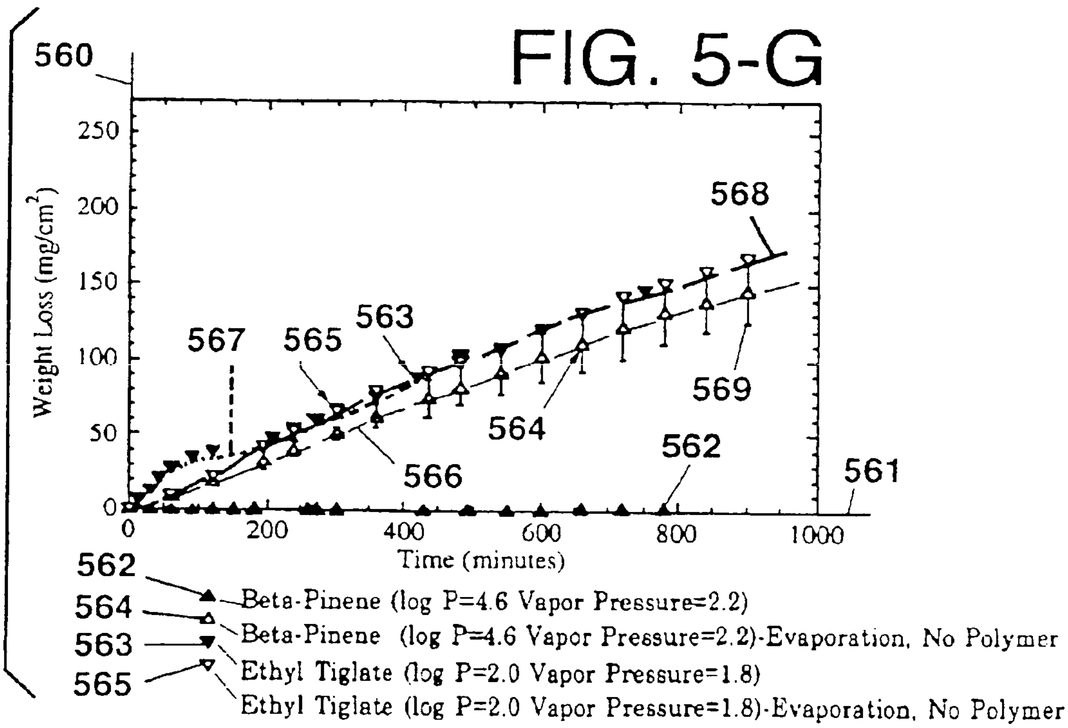

560
250
200
150
100
50
0
Weight Loss (mg/cm^2)
568
563
567
565
569
564
562
566
561
0
200
400
600
800
1000
Time (minutes)
562
564
563
565
Beta-Pinene (log P=4.6 Vapor Pressure=2.2)
Beta-Pinene (log P=4.6 Vapor Pressure=2.2)-Evaporation, No Polymer
Ethyl Tiglate (log P=2.0 Vapor Pressure=1.8)
Ethyl Tiglate (log P=2.0 Vapor Pressure=1.8)-Evaporation, No Polymer

FIG. 5-H

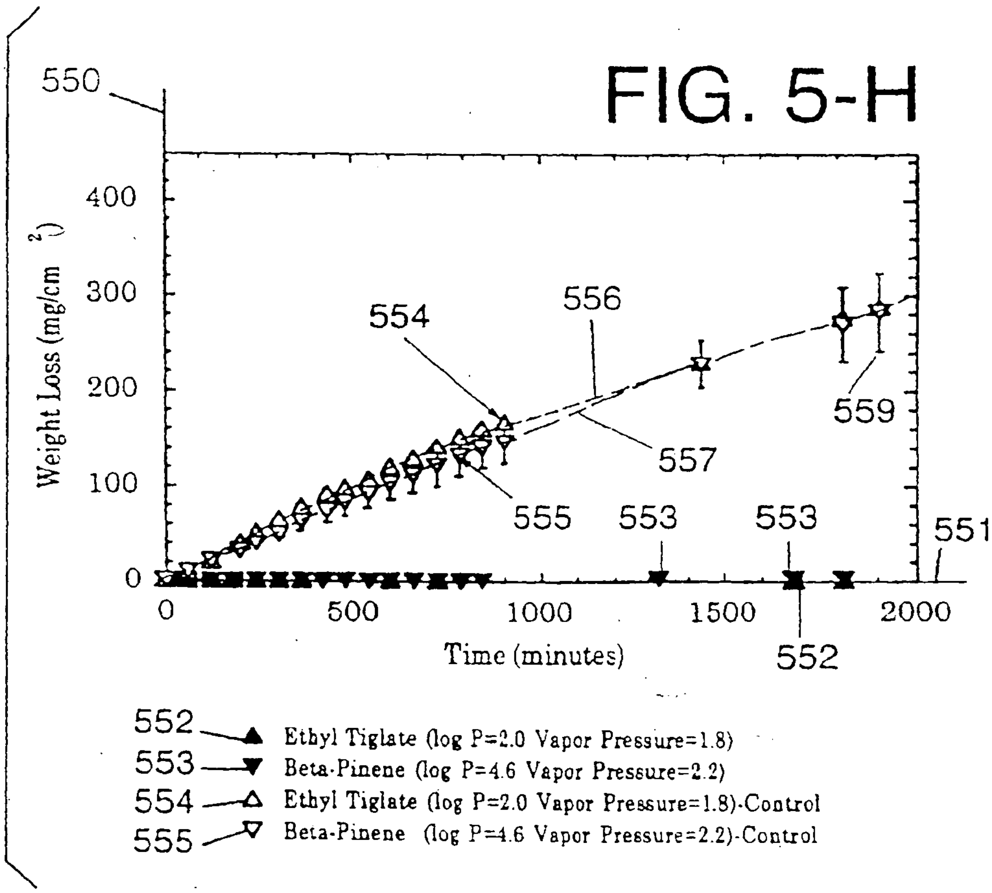
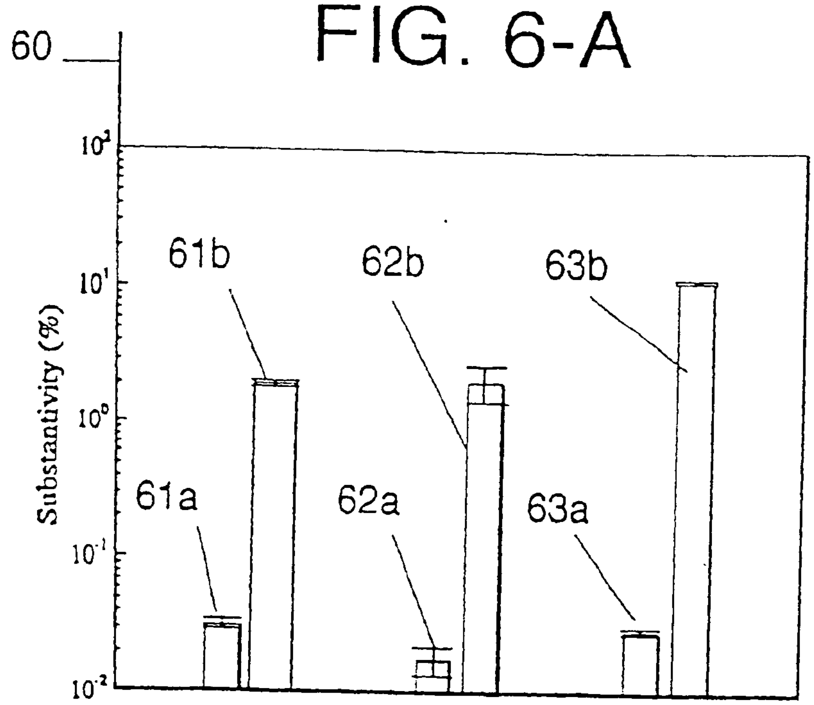

550
551
554
556
557
555
553
553
552
559
400
300
200
100
0
Weight Loss (mg/cm²)
0
500
1000
1500
2000
Time (minutes)
552 Ethyl Tiglate (log P=2.0 Vapor Pressure=1.8)
553 Beta-Pinene (log P=4.6 Vapor Pressure=2.2)
554 Ethyl Tiglate (log P=2.0 Vapor Pressure=1.8)-Control
555 Beta-Pinene (log P=4.6 Vapor Pressure=2.2)-Control

FIG. 6-A


60
10^2
10^1
10^0
10^{-1}
10^{-2}
Substantivity (%)
61b
62b
63b
61a
62a
63a

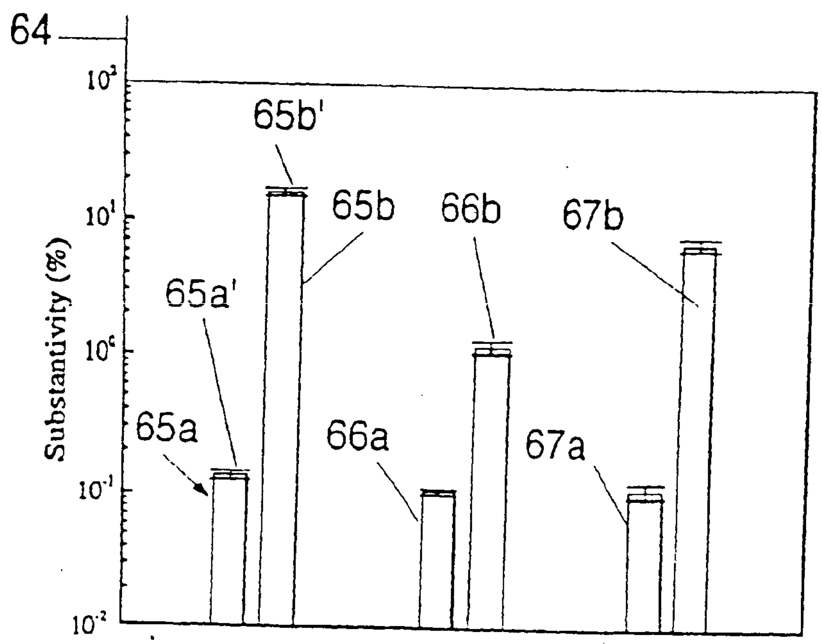

64
10^2
10^1
10^0
10^{-1}
10^{-2}
Substantivity (%)
65b'
65b
66b
67b
65a'
65a
66a
67a

FIG. 6-B

FIG. 6-C

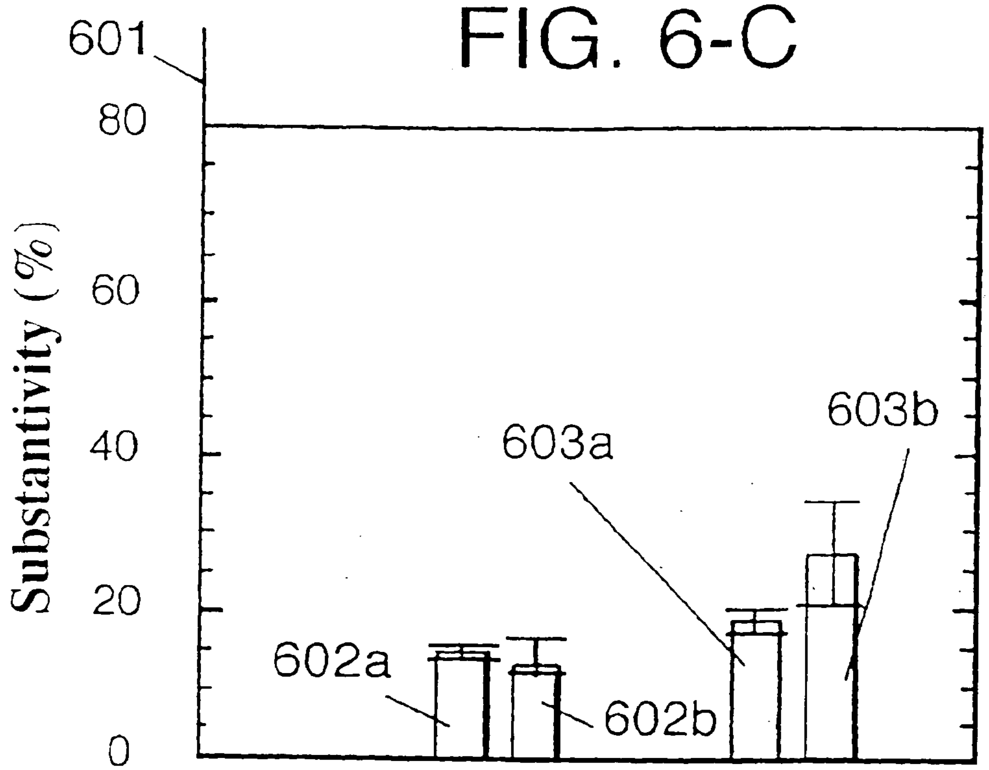

601
80
60
40
20
0
Substantivity (%)
603a
603b
602a
602b

FIG. 6-D

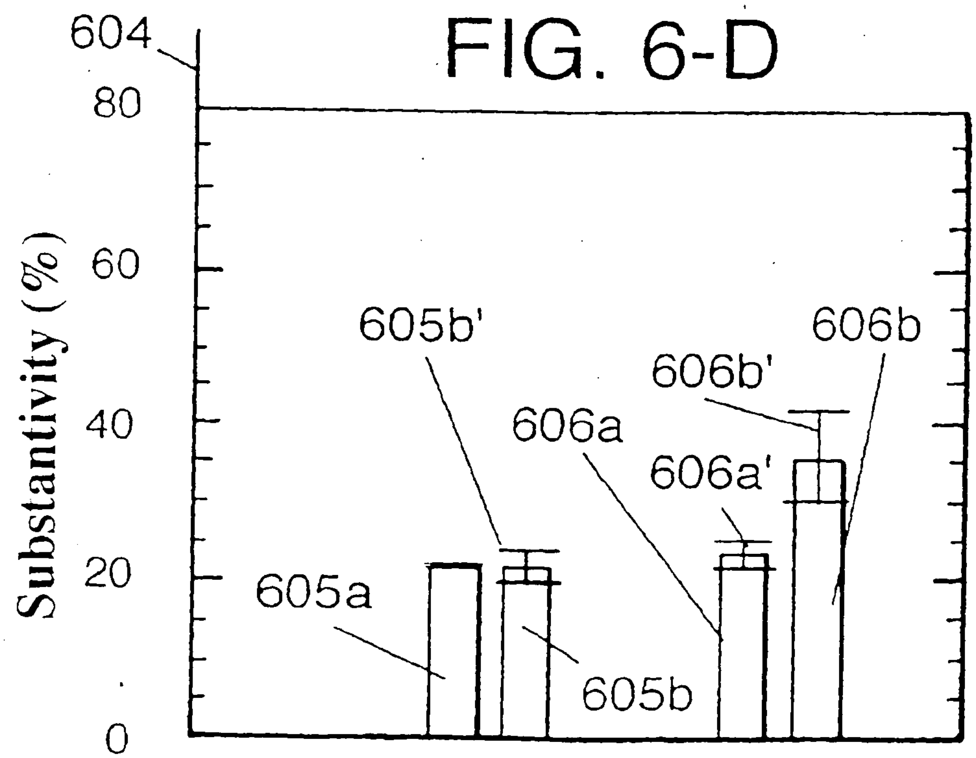

604
80
60
40
20
0
Substantivity (%)
605b'
606b
606b'
606a
606a'
605a
605b

FIG. 7-A

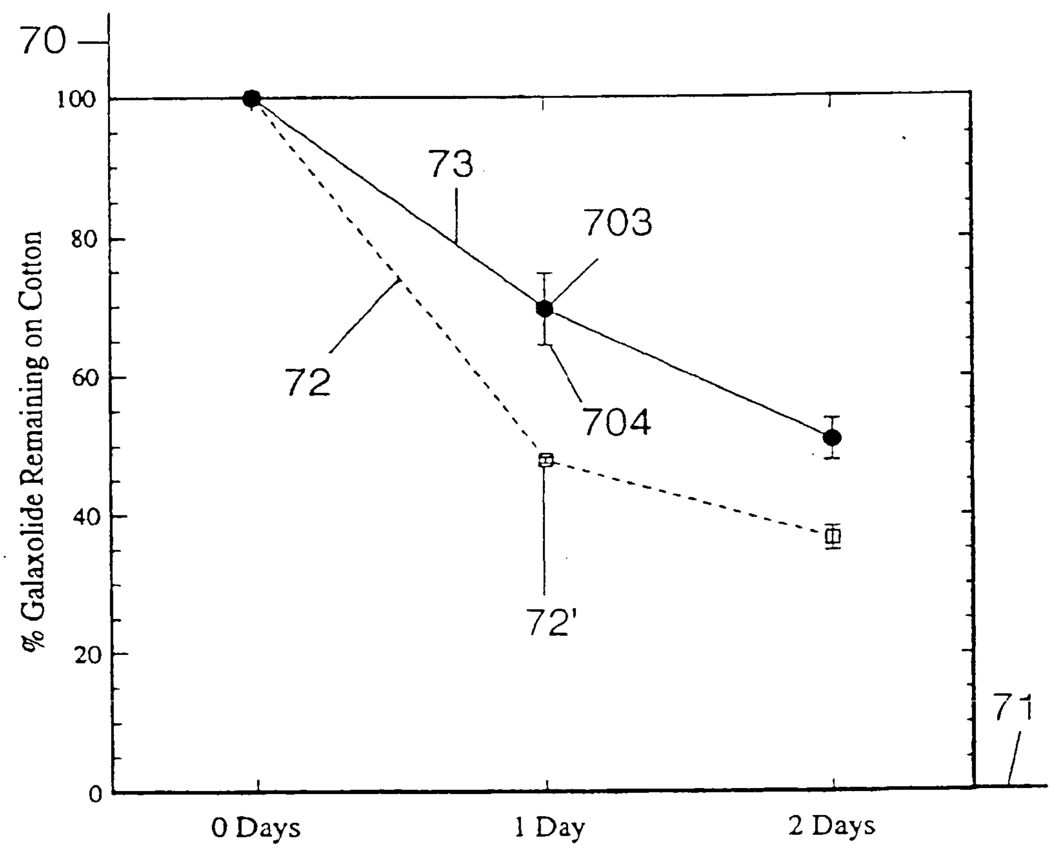

70
100
80
60
40
20
0
% Galaxolide Remaining on Cotton
73
703
72
704
72'
71
0 Days
1 Day
2 Days

FIG. 7-B

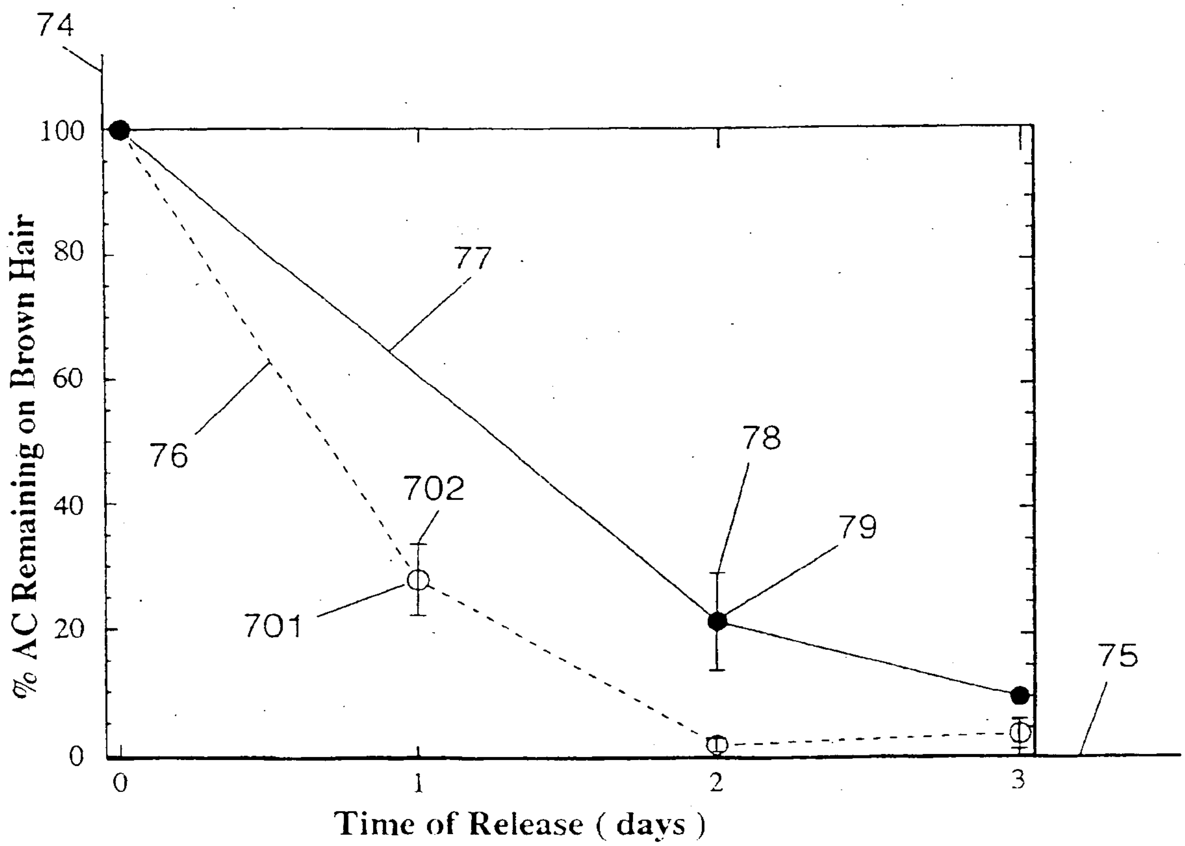
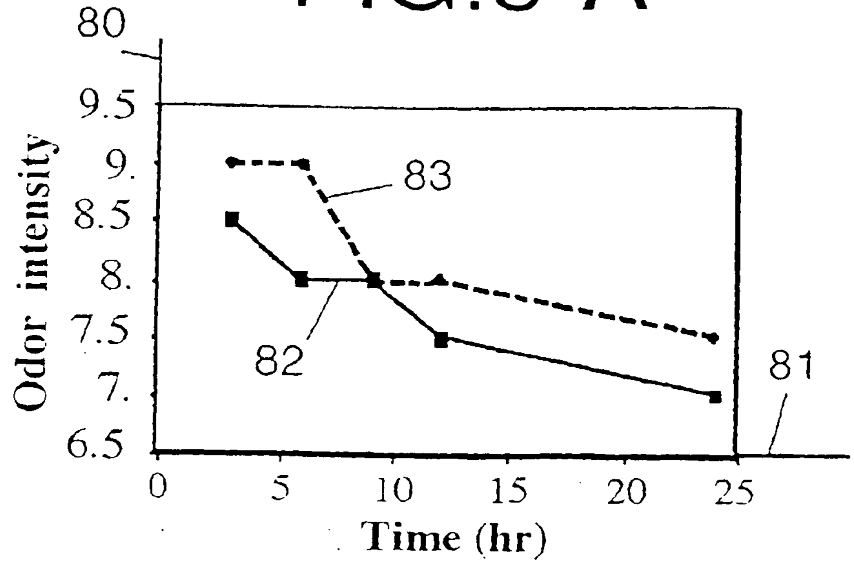

74
100
80
60
40
20
0
% AC Remaining on Brown Hair
77
76
702
78
79
701
75
0
1
2
3
Time of Release (days)

FIG.8-A


80
9.5
9.
8.5
8.
7.5
7.
6.5
Odor intensity
83
82
81
0
5
10
15
20
25
Time (hr)

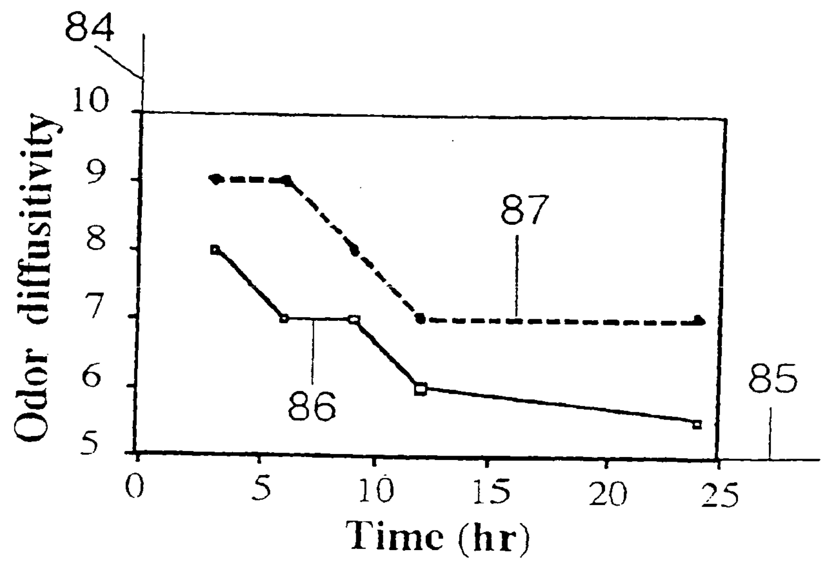

84
10
9
8
7
6
5
Odor diffusivity
87
86
85
0
5
10
15
20
25
Time (hr)

FIG.8-B

FIG. 8-C

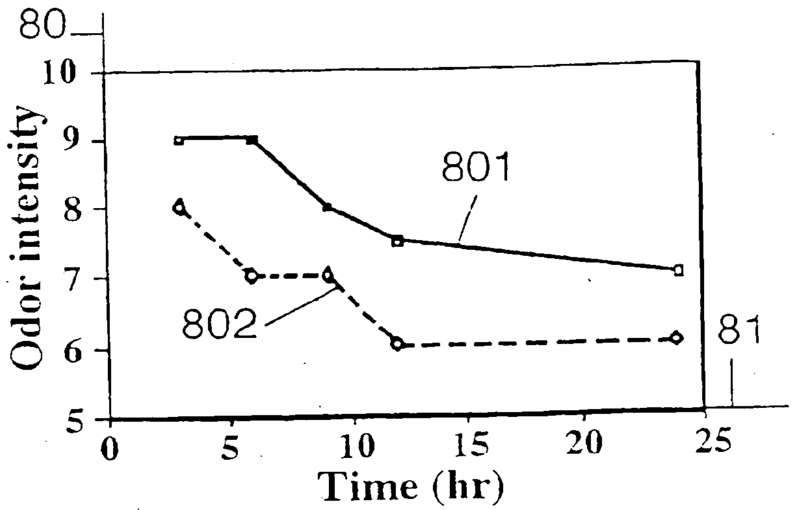

80
10
9
8
7
6
5
Odor intensity
801
802
81
0
5
10
15
20
25
Time (hr)

FIG. 8-D

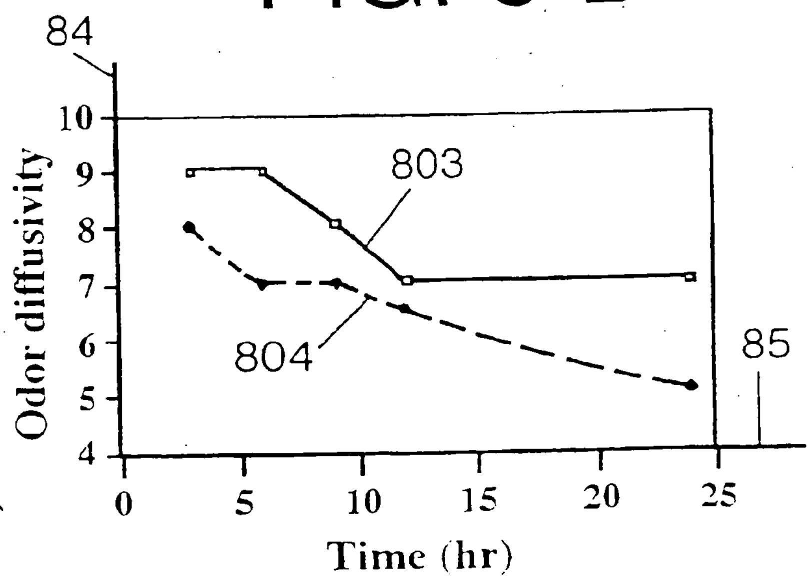

84
10
9
8
7
6
5
4
Odor diffusivity
803
804
85
0
5
10
15
20
25
Time (hr)

FIG.8-E

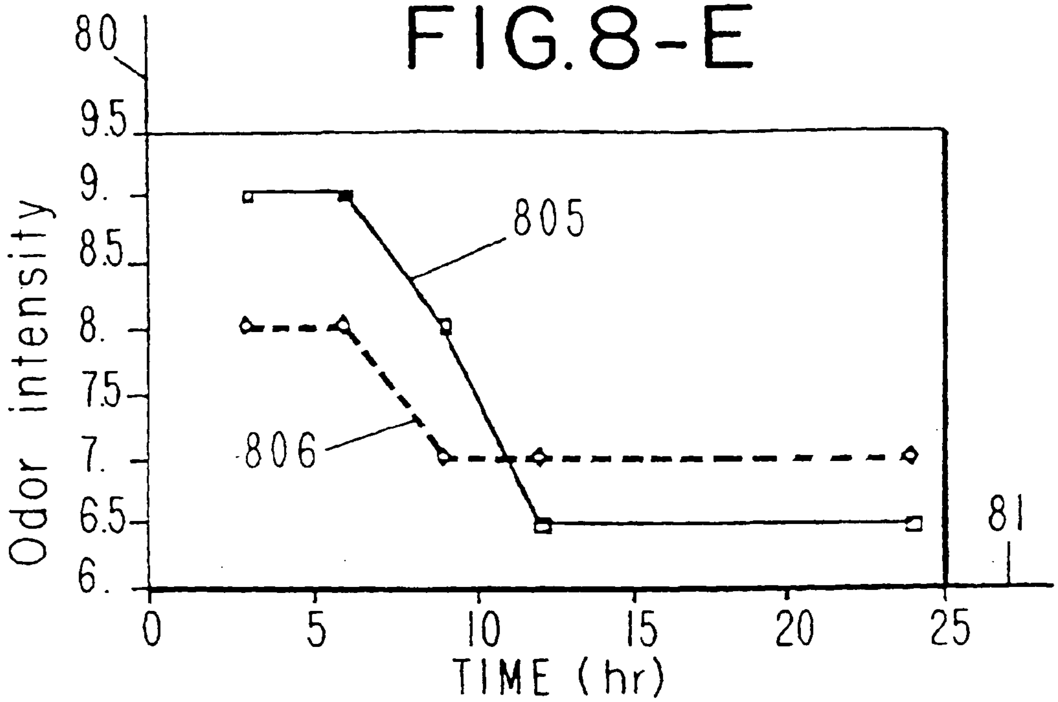

80
9.5
9.
8.5
8.
7.5
7.
6.5
6.
Odor intensity
805
806
81
0
5
10
15
20
25
TIME (hr)

FIG.8-F

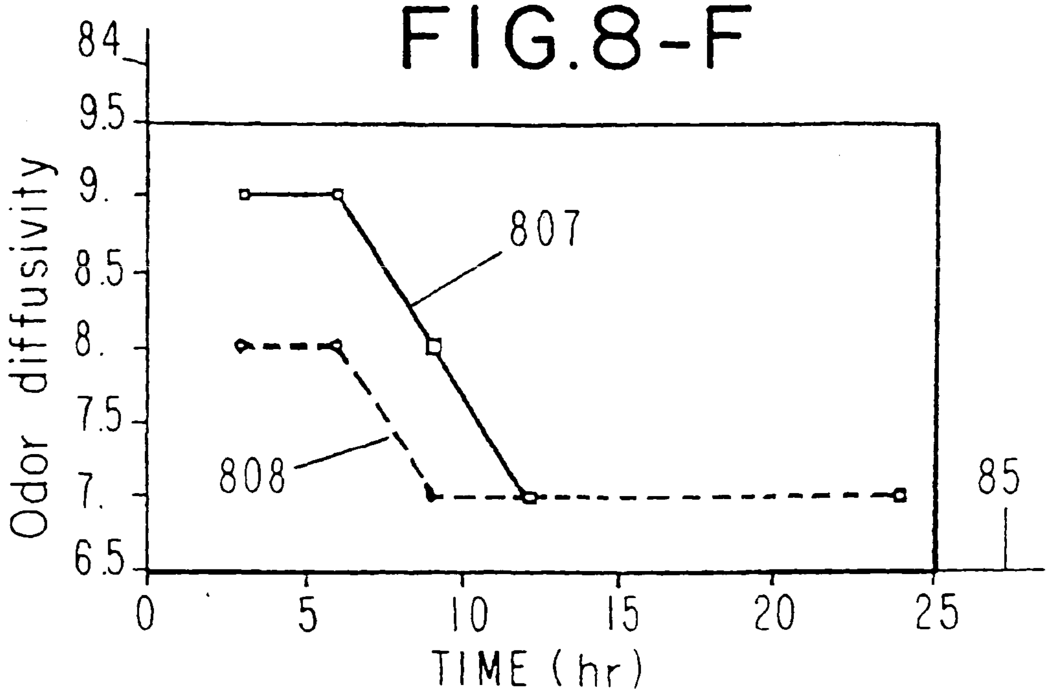

84
9.5
9.
8.5
8.
7.5
7.
6.5
Odor diffusivity
807
808
85
0
5
10
15
20
25
TIME (hr)

FIG. 8-G

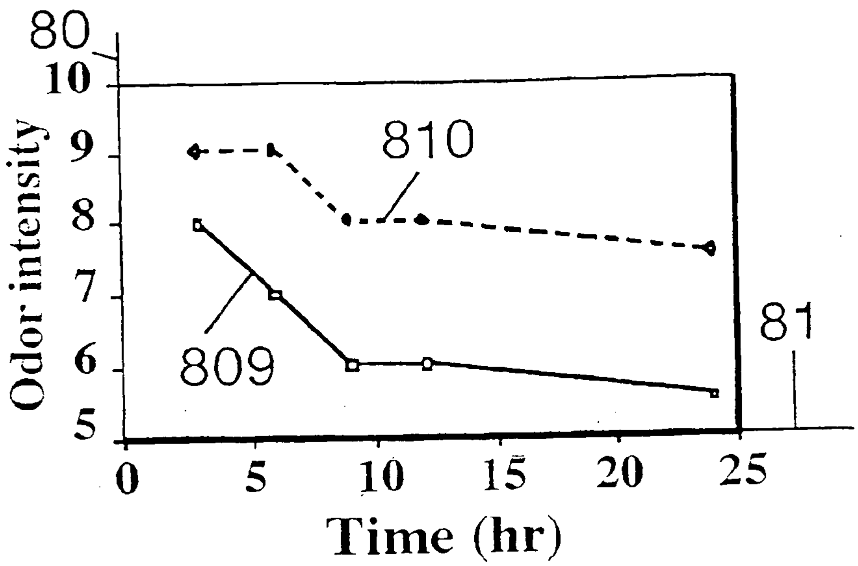

80
81
809
810
Odor intensity
10
9
8
7
6
5
0
5
10
15
20
25
Time (hr)

FIG. 8-H

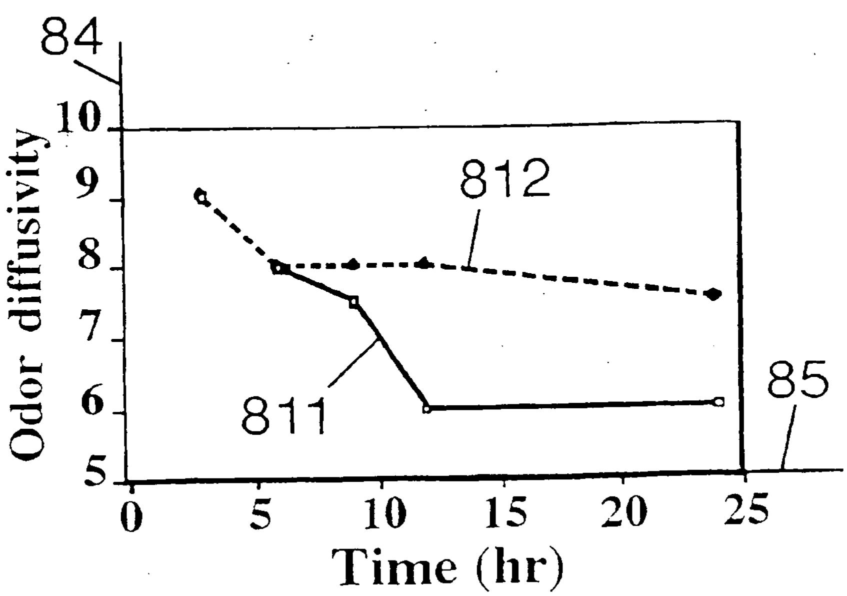

84
85
811
812
Odor diffusivity
10
9
8
7
6
5
0
5
10
15
20
25
Time (hr)

FIG.8-I

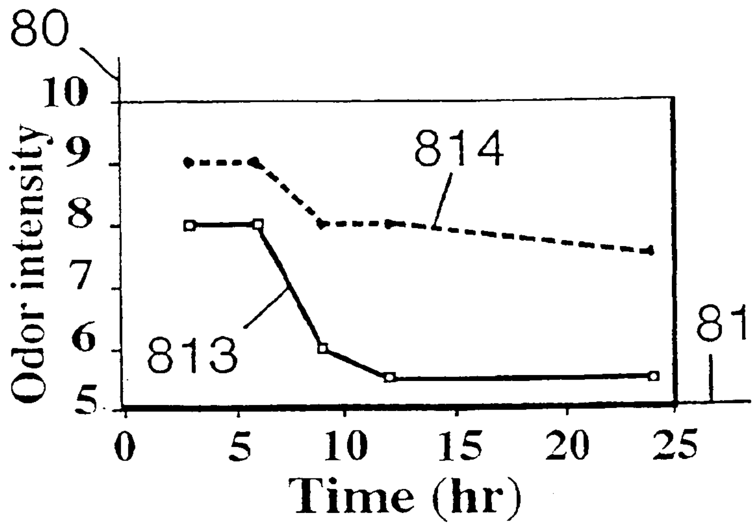

80
10
9
8
7
6
5
Odor intensity
814
813
81
0
5
10
15
20
25
Time (hr)

FIG.8-J

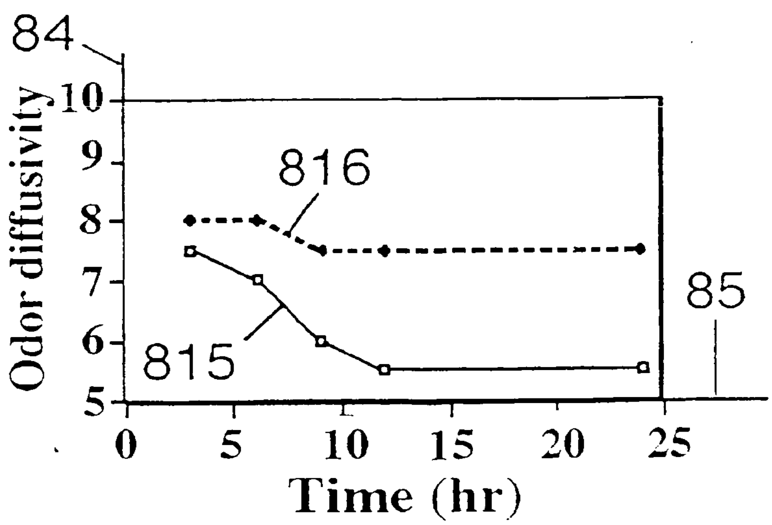
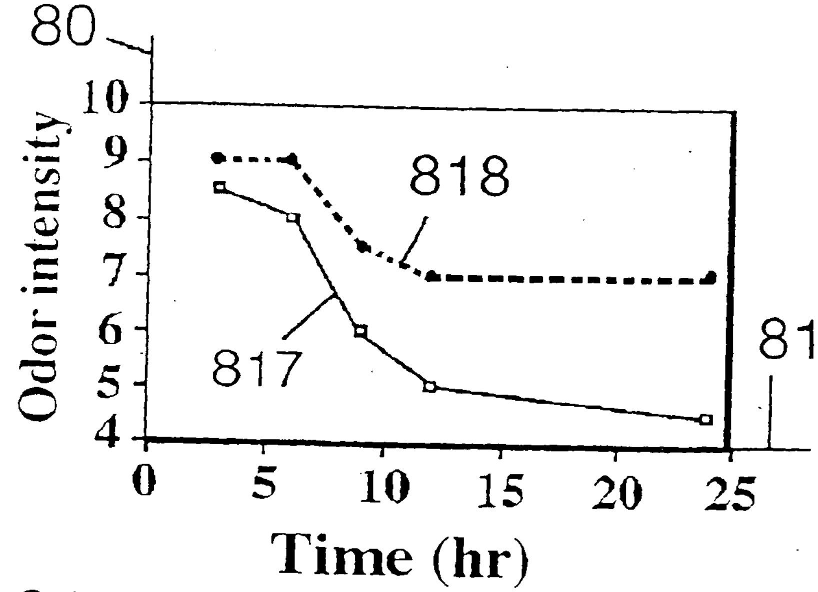

84
10
9
8
7
6
5
Odor diffusivity
816
815
85
0
5
10
15
20
25
Time (hr)

FIG. 8-K


80
10
9
8
7
6
5
4
Odor intensity
818
817
81
0
5
10
15
20
25
Time (hr)

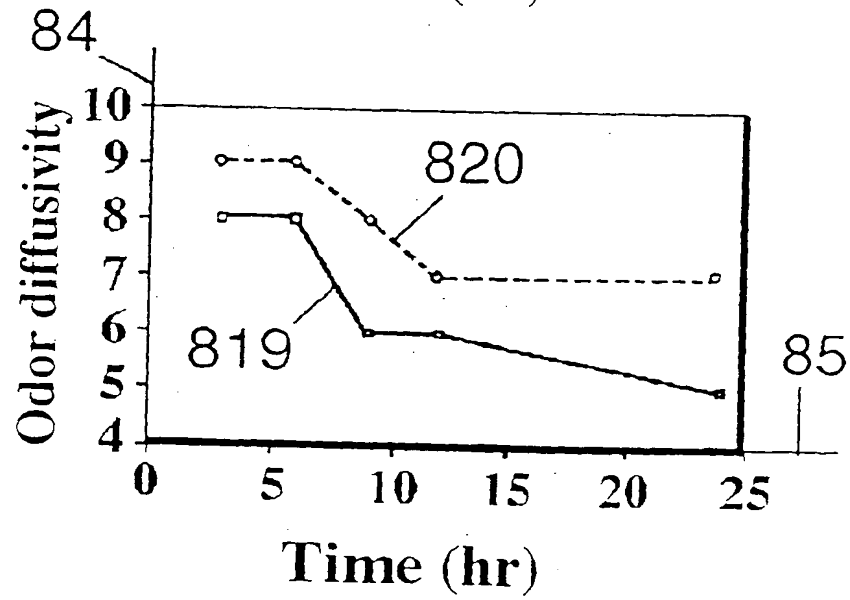
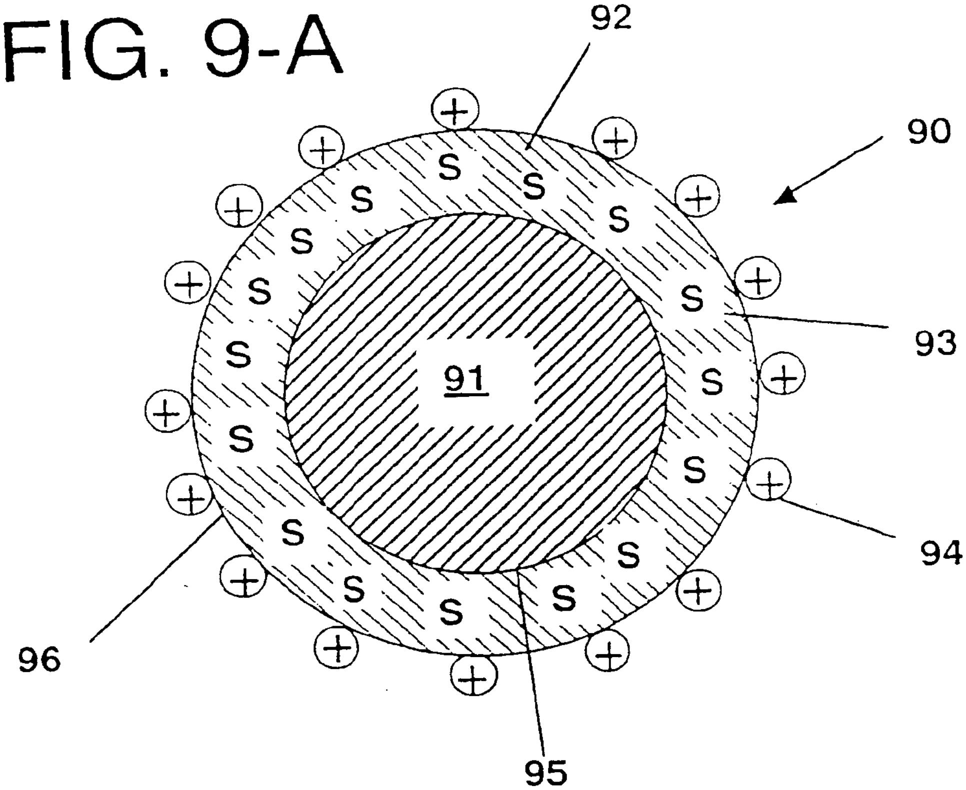

84
10
9
8
7
6
5
4
Odor diffusivity
820
819
85
0
5
10
15
20
25
Time (hr)

FIG. 8-L

FIG. 9-A



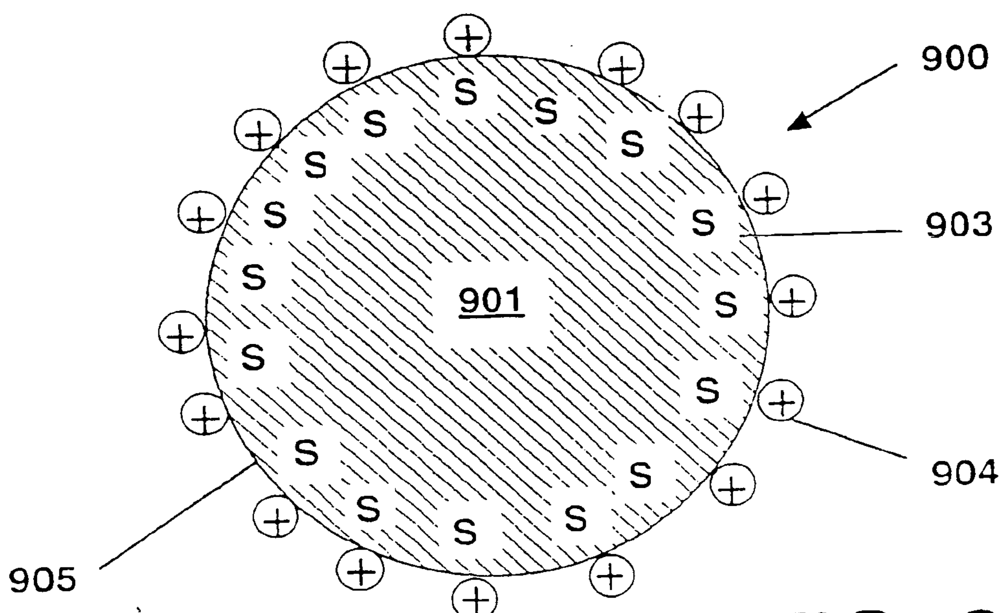
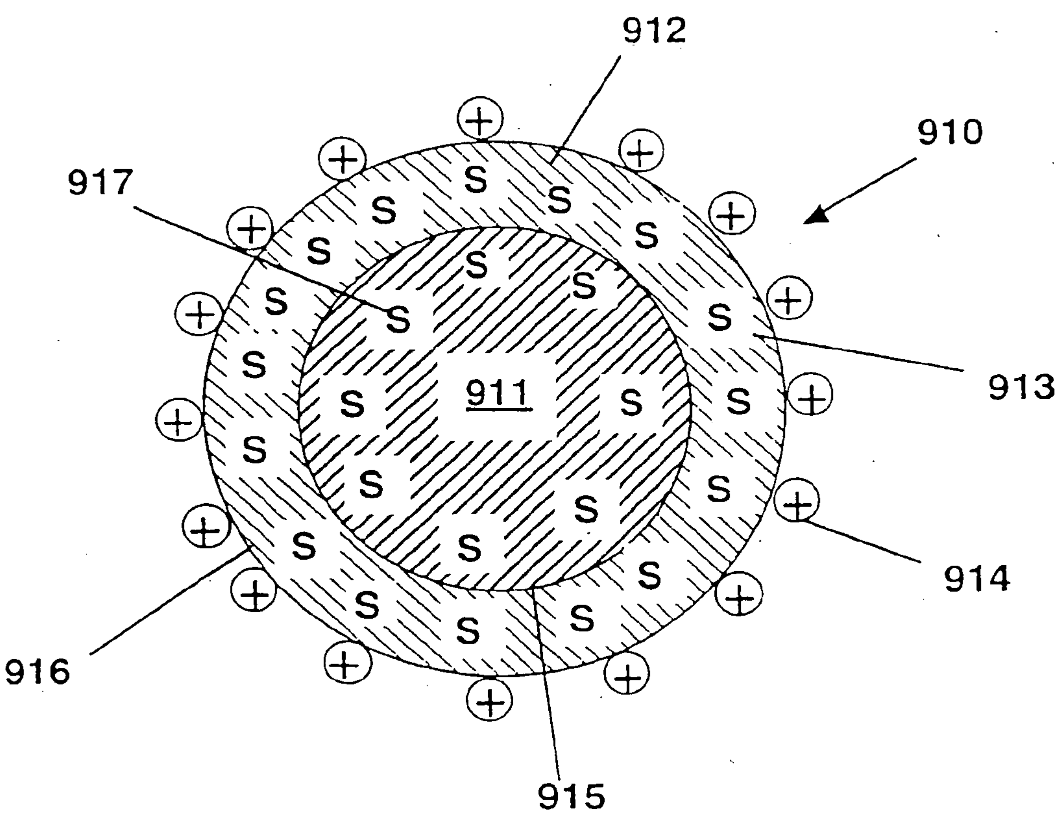


FIG. 9-B

FIG. 9-C



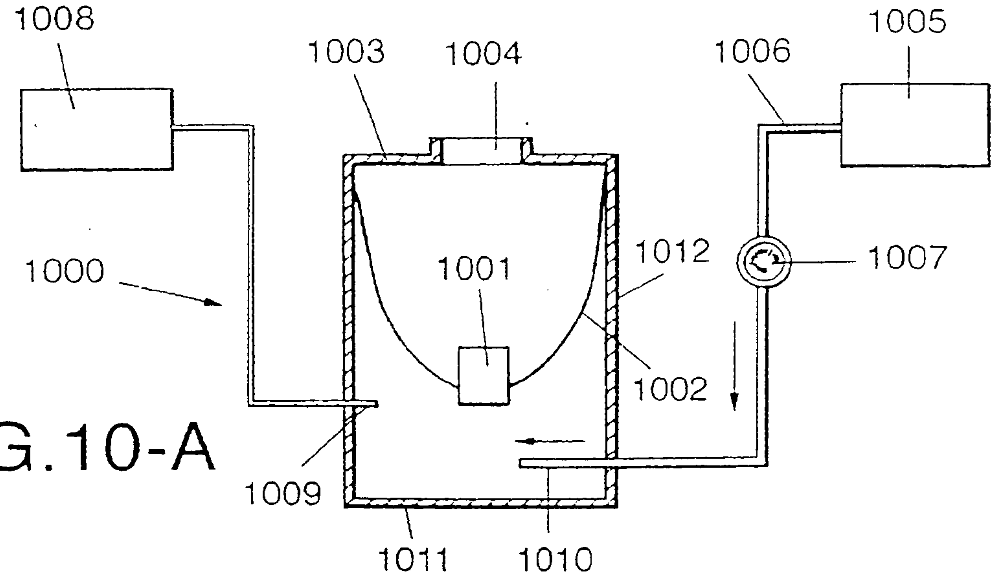


FIG.10-A

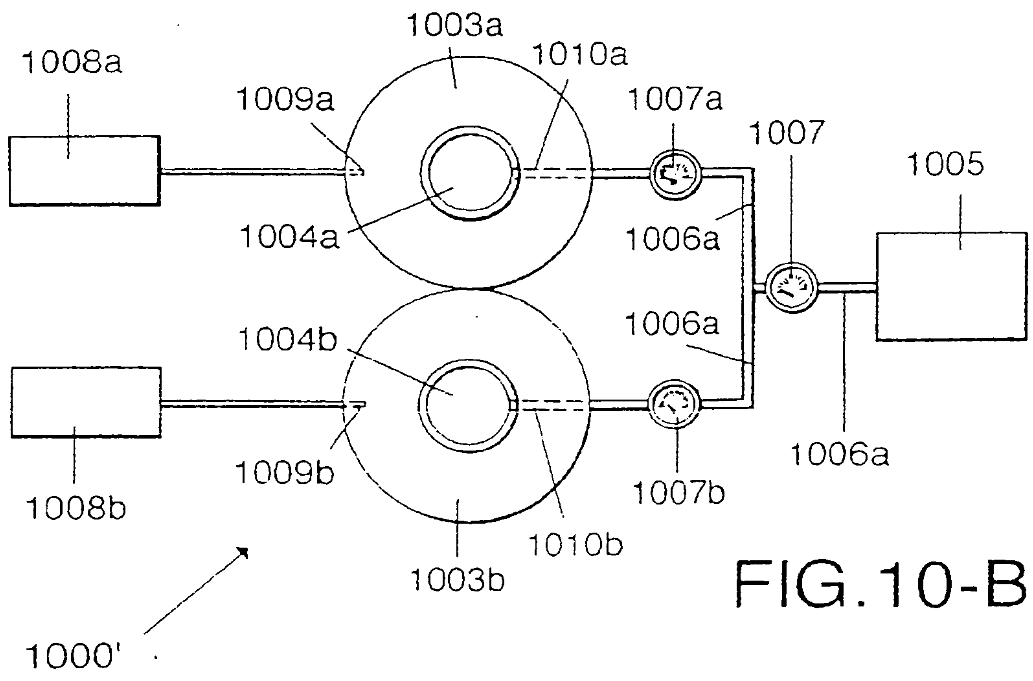
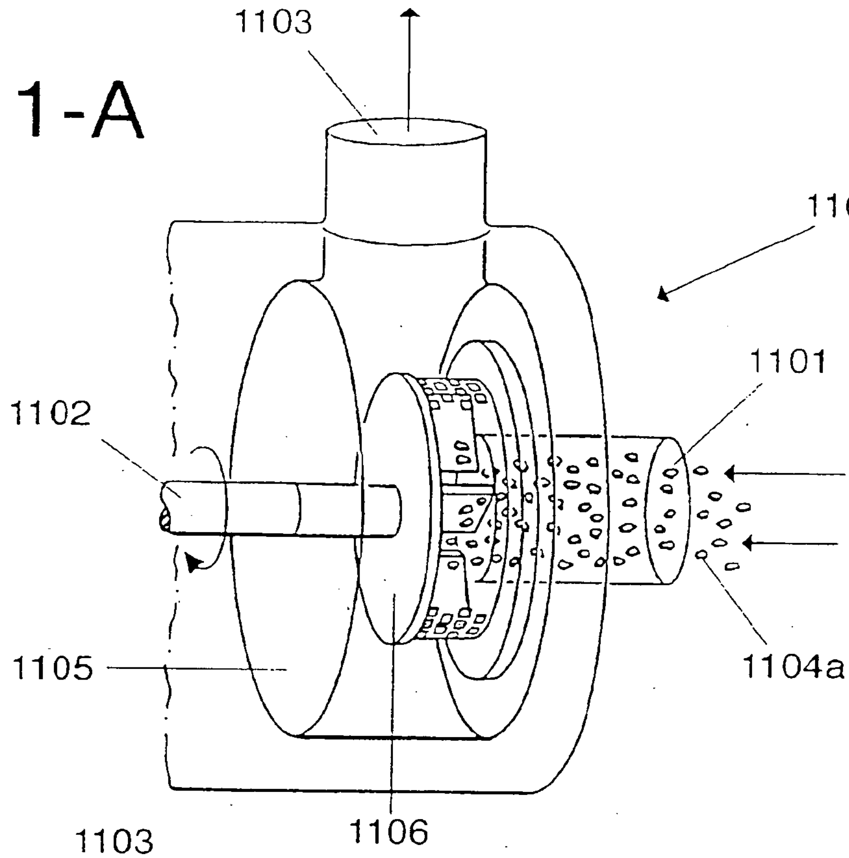


FIG.10-B

FIG.11-A



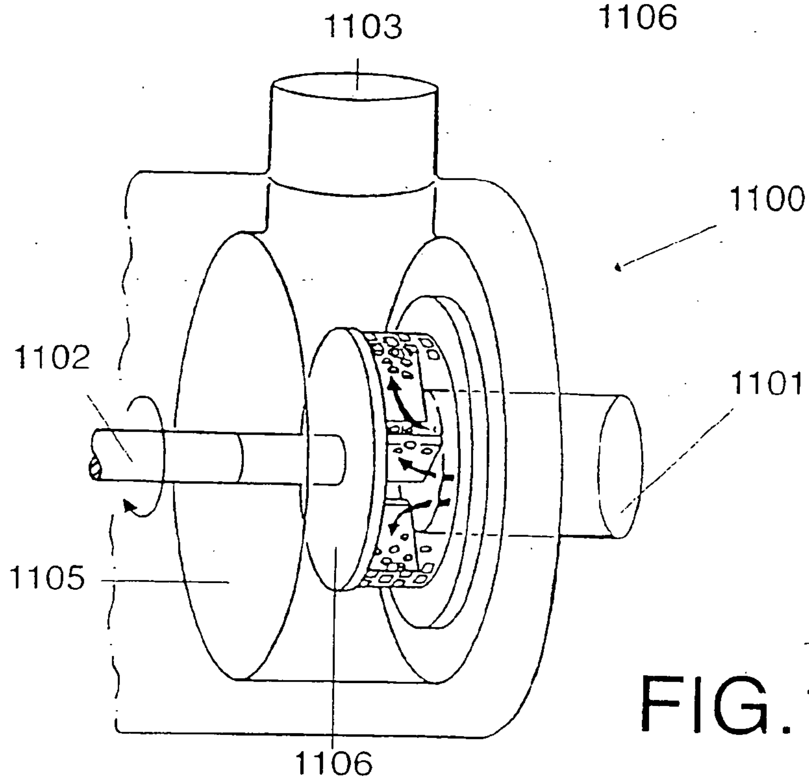
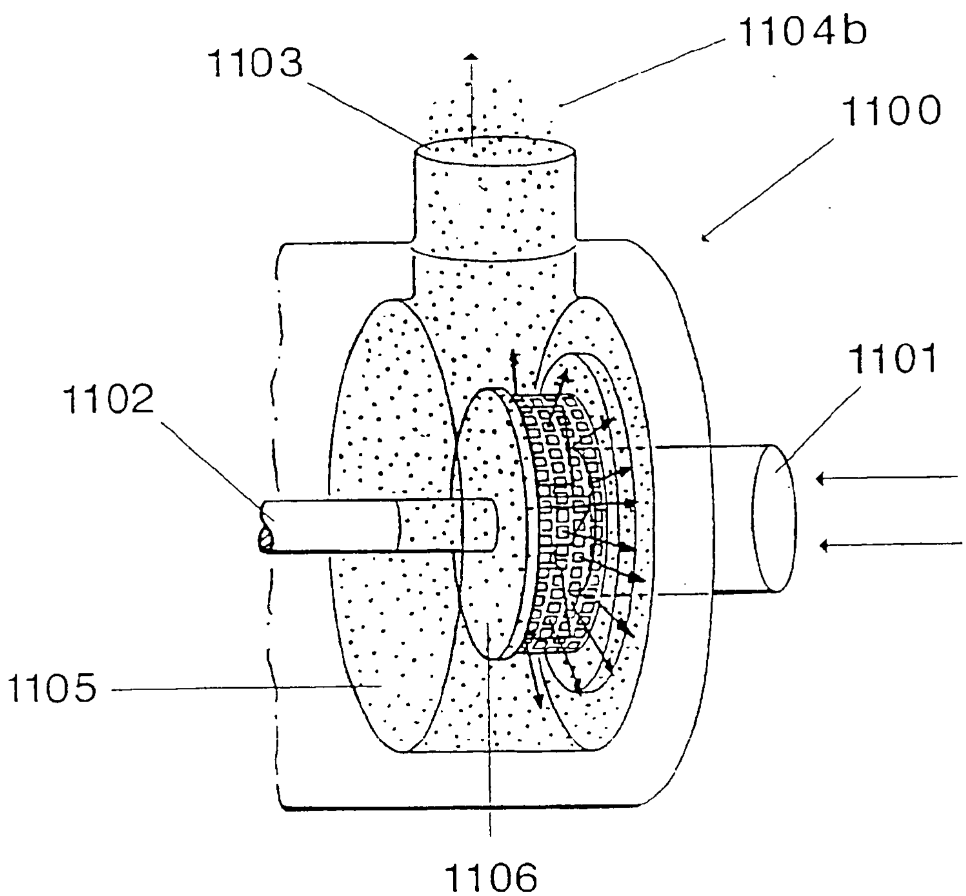


FIG.11-B

FIG. 11-C



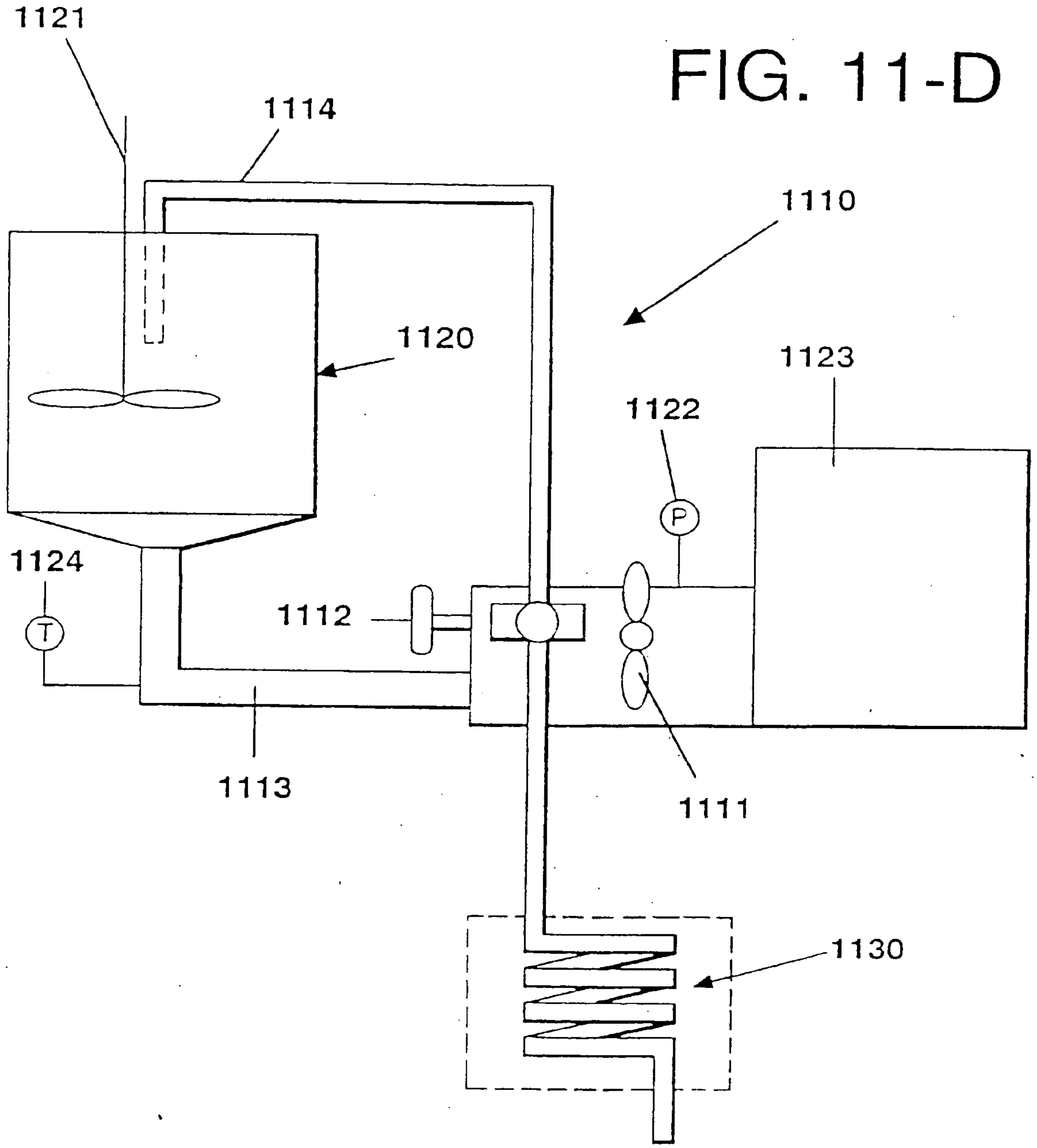


FIG. 11-D

FIG.11-E

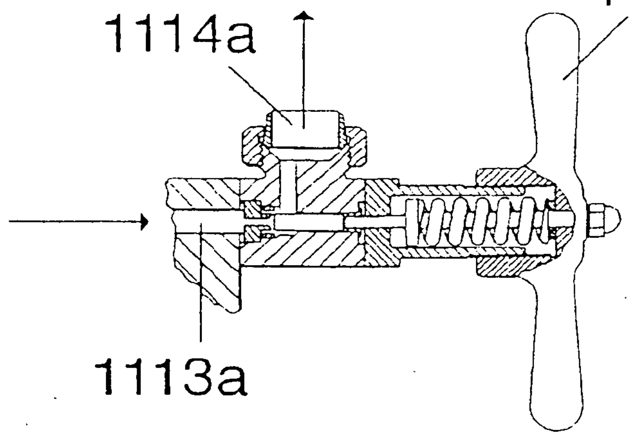
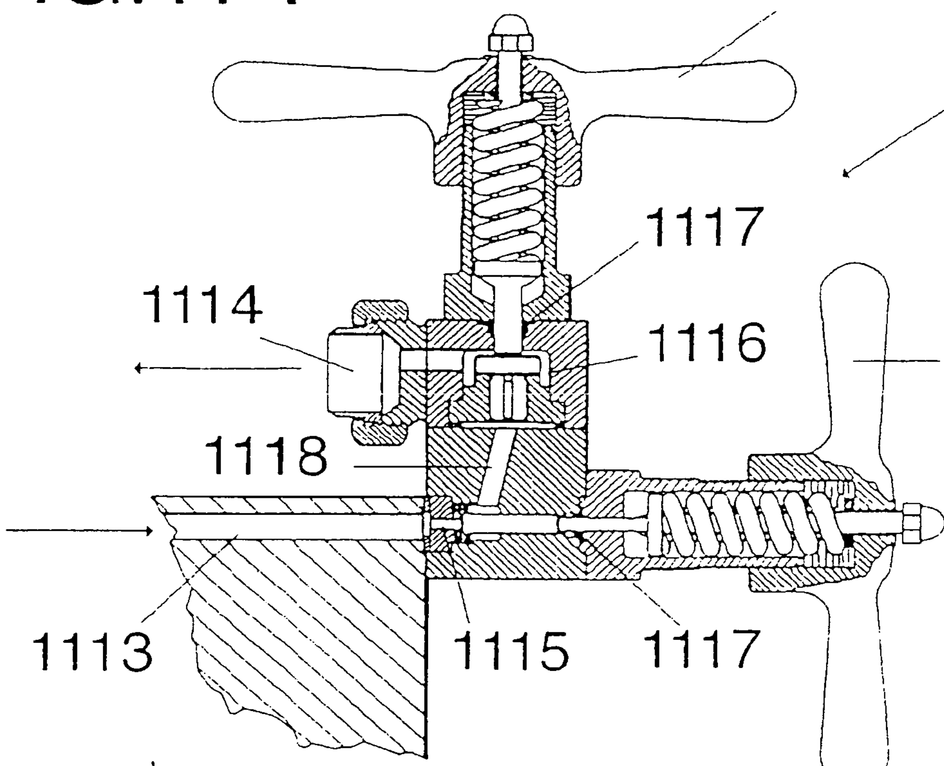


FIG.11-F



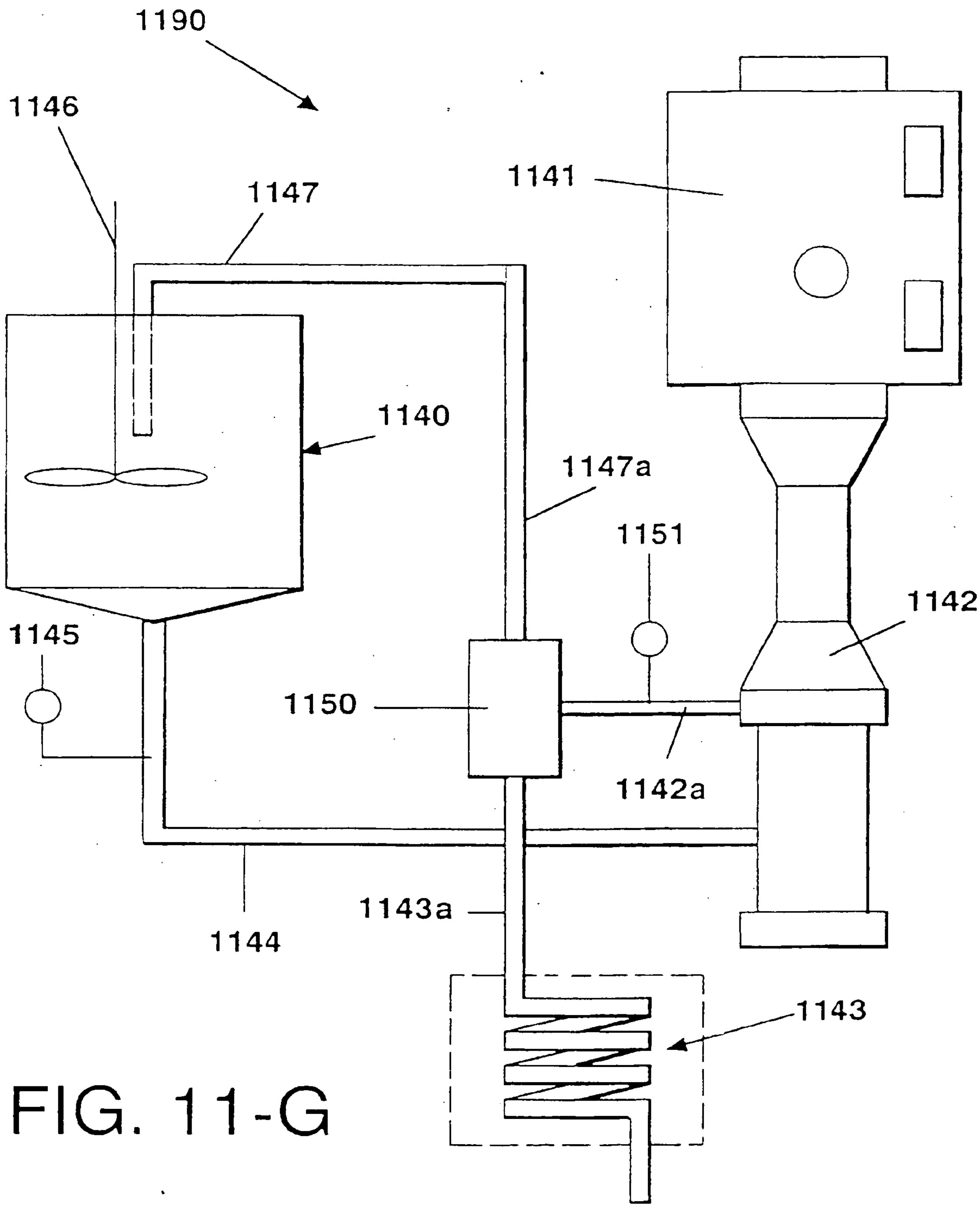

1190
1146
1147
1141
1140
1147a
1151
1142
1145
1150
1142a
1143a
1144
1143

FIG. 11-G

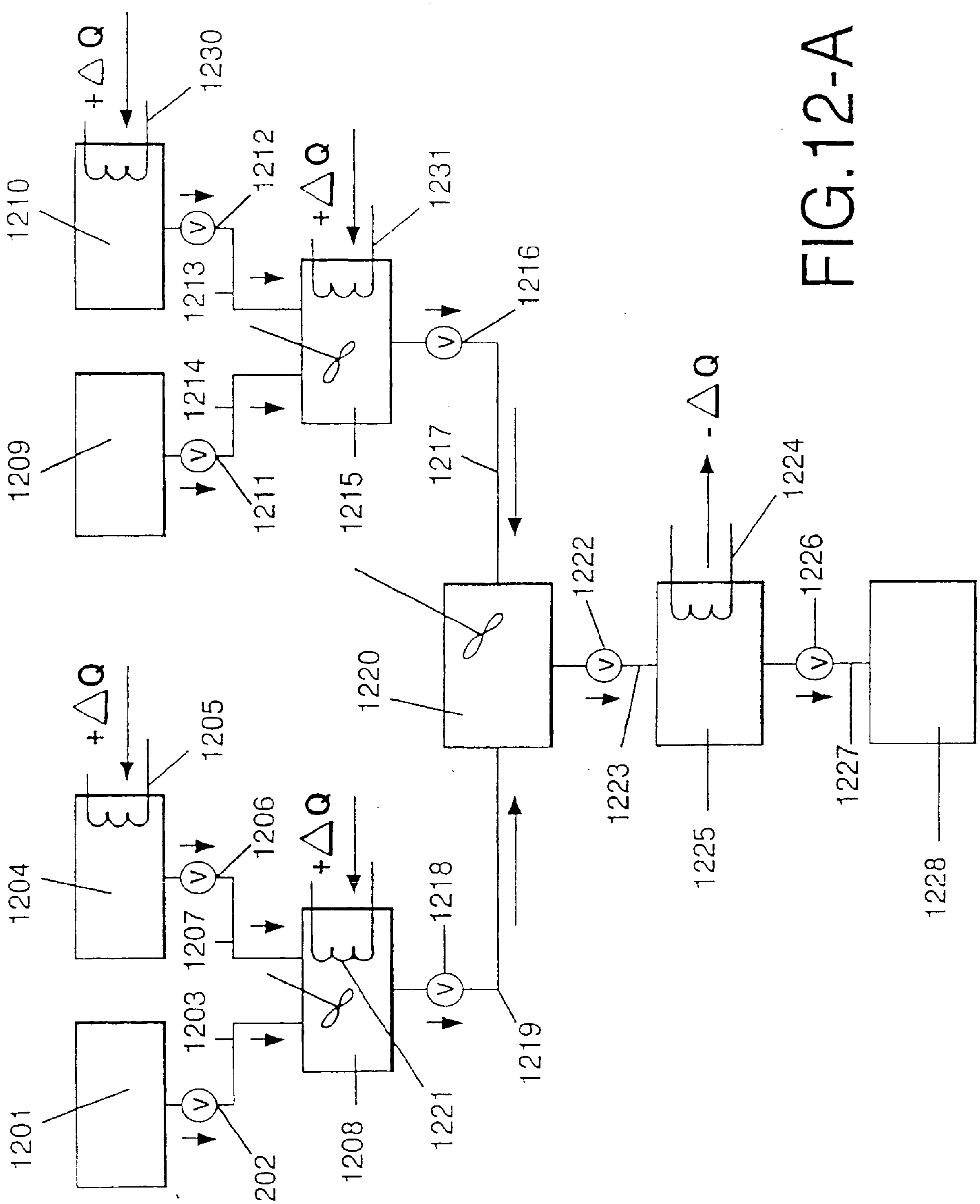

1201
1202
1203
1204
1205
1206
1207
1208
1209
1210
1211
1212
1213
1214
1215
1216
1217
1218
1219
1220
1221
1222
1223
1224
1225
1226
1227
1228
1230
1231
+ΔQ
+ΔQ
+ΔQ
+ΔQ
- ΔQ
V

FIG.12-A

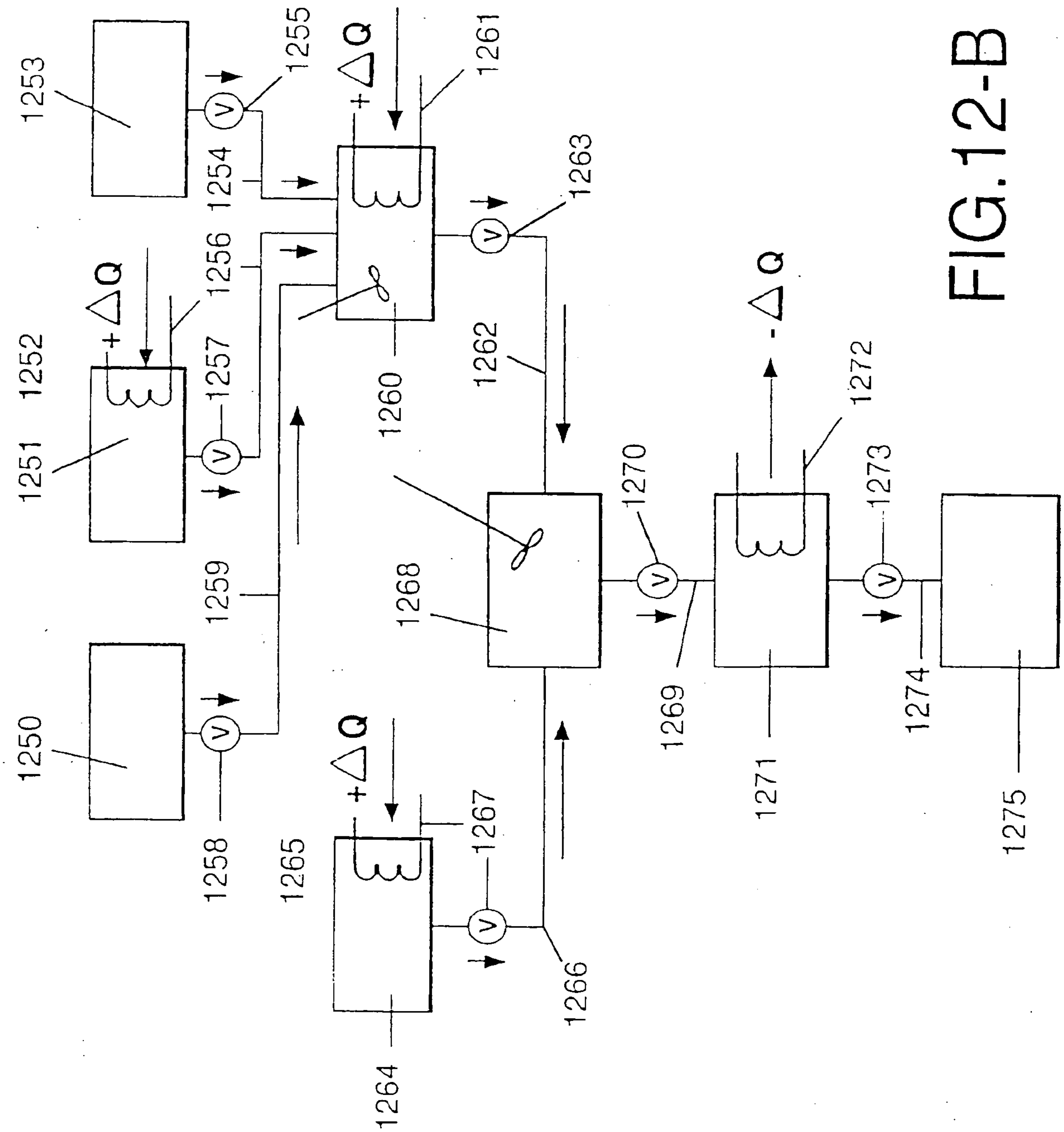
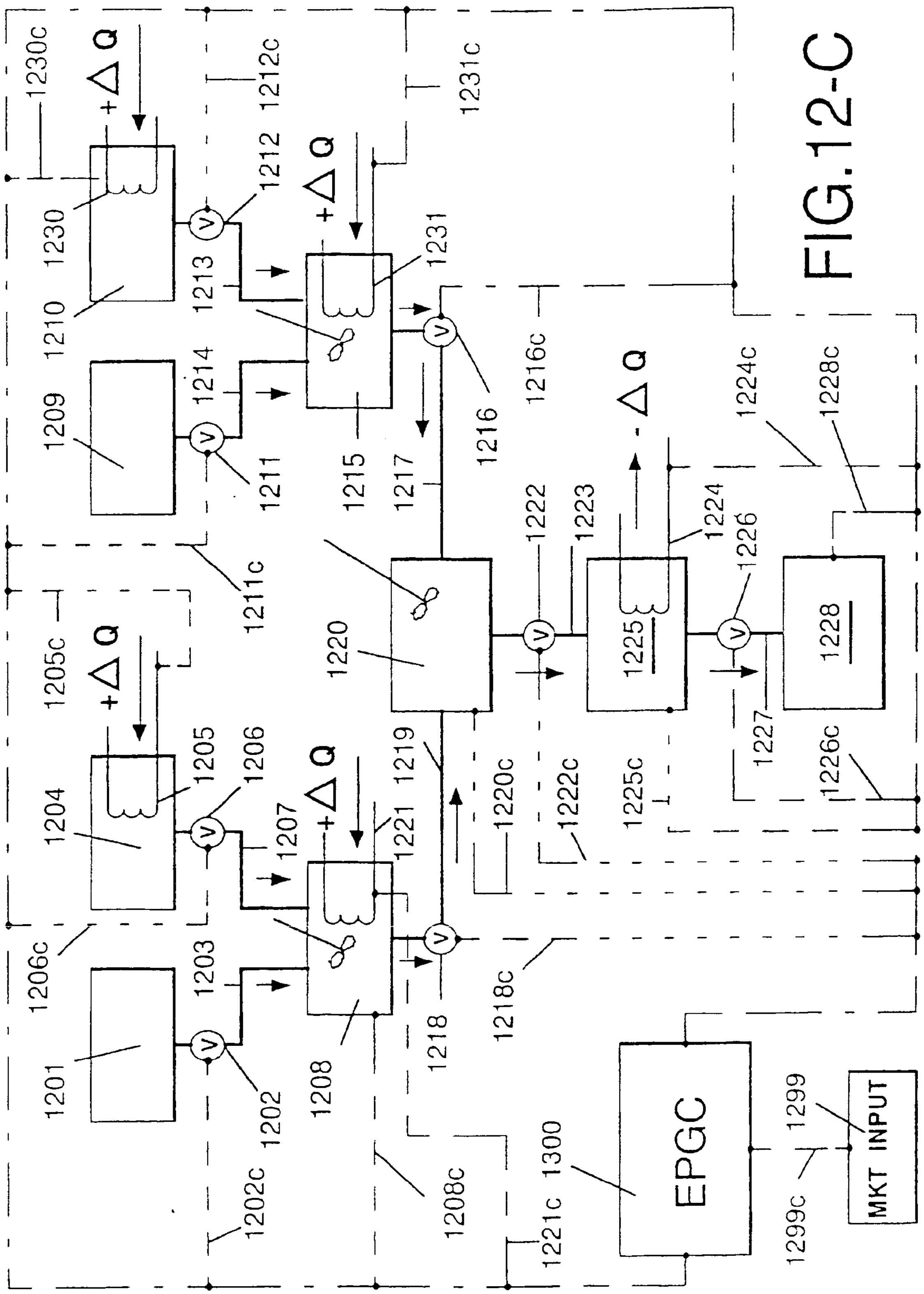

1250
1251
1252
1253
1254
1255
1256
1257
1258
1259
1260
1261
1262
1263
1264
1265
1266
1267
1268
1269
1270
1271
1272
1273
1274
1275
+ΔQ
+ΔQ
+ΔQ
- ΔQ
V

FIG.12-B



1230c
+ΔQ
1212c
1231c
1230
1212
+ΔQ
1210
1213
1231
1209
1214
1216c
-ΔQ
1224c
1228c
1211
1215
1217
1216
1222
1223
1224
1226
1211c
1220
1225
1228
1205c
+ΔQ
1205
1206
1204
1207
+ΔQ
1221
1219
1220c
1222c
1225c
1227
1226c
1206c
1203
1218c
1201
1202
1208
1218
EPGC
1300
1299
MKT INPUT
1202c
1208c
1221c
1299c

FIG.12-C

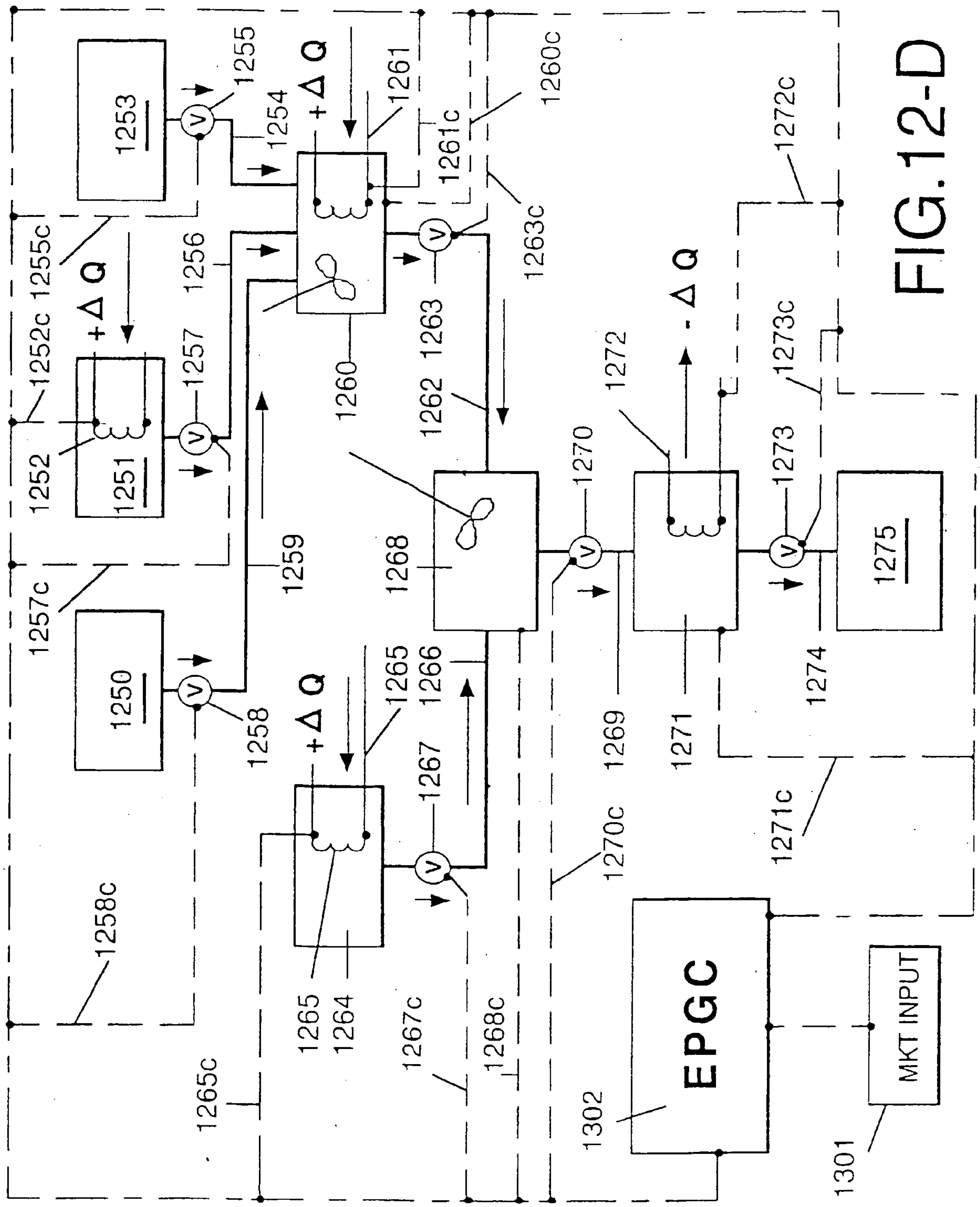

1253
1255
1254
+ΔQ
1261
1261c
1260c
1263c
1255c
1252c
+ΔQ
1256
1257
1263
1260
1262
1252
1251
1259
1268
1270
1272
-ΔQ
1272c
1273c
1273
1275
1257c
1250
1258
+ΔQ
1265
1266
1267
1269
1271
1274
1271c
1270c
1258c
1265
1264
1267c
1268c
1265c
EPGC
1302
MKT INPUT
1301

FIG.12-D

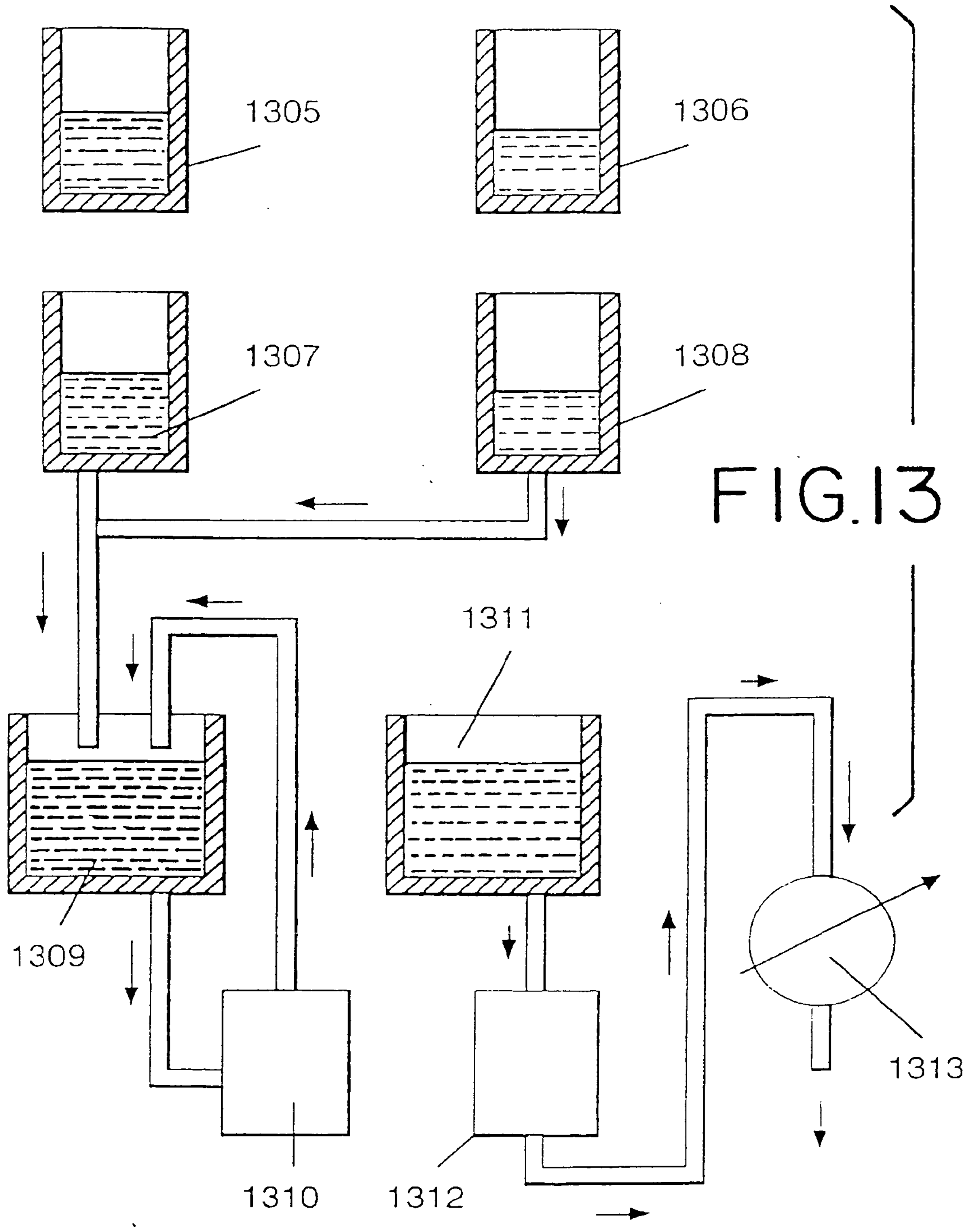


FIG.13

FIG.14

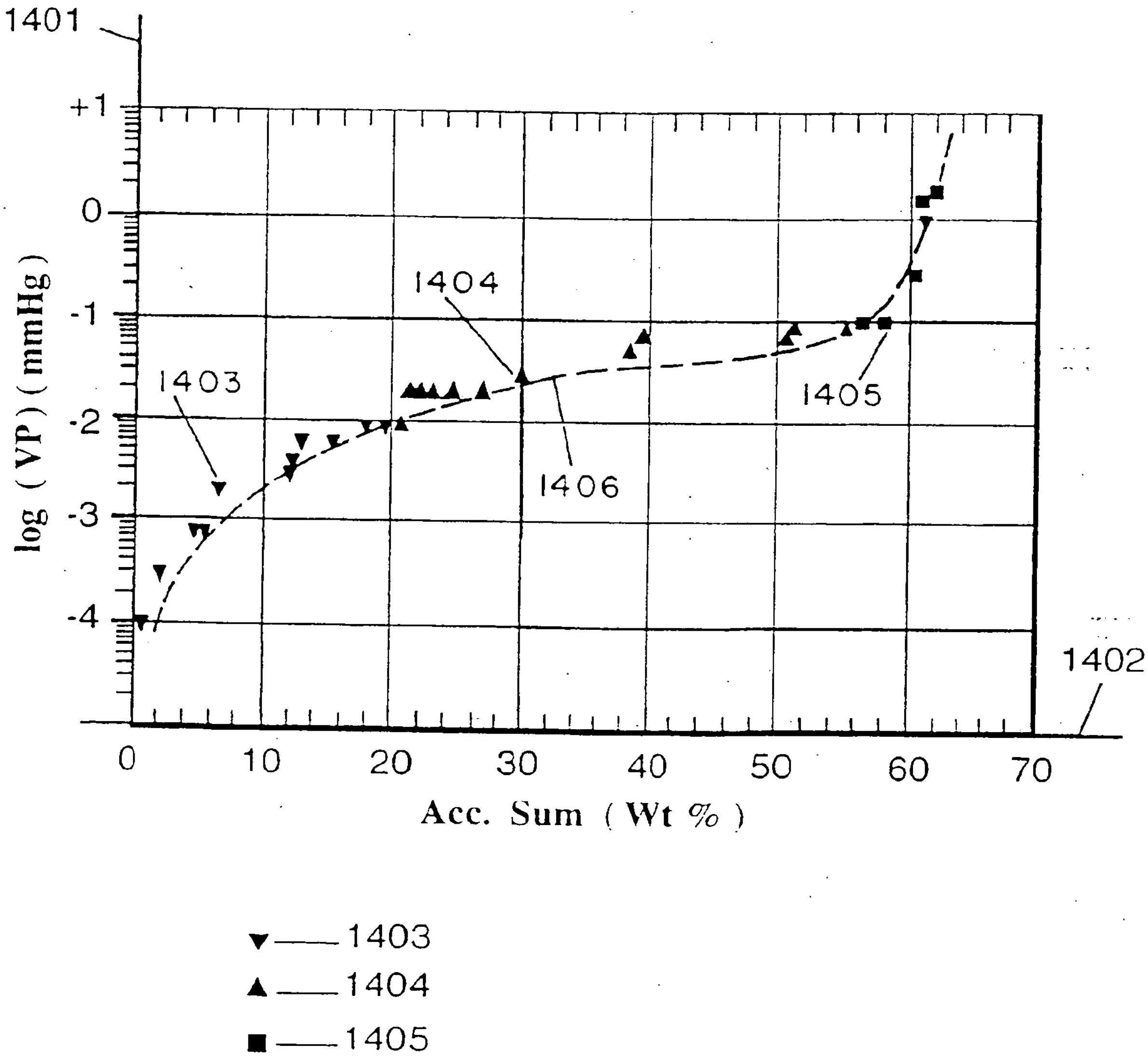

1401
+1
0
-1
-2
-3
-4
log (VP) (mmHg)
1404
1403
1405
1406
1402
0
10
20
30
40
50
60
70
Acc. Sum (Wt %)
1403
1404
1405

FIG.15

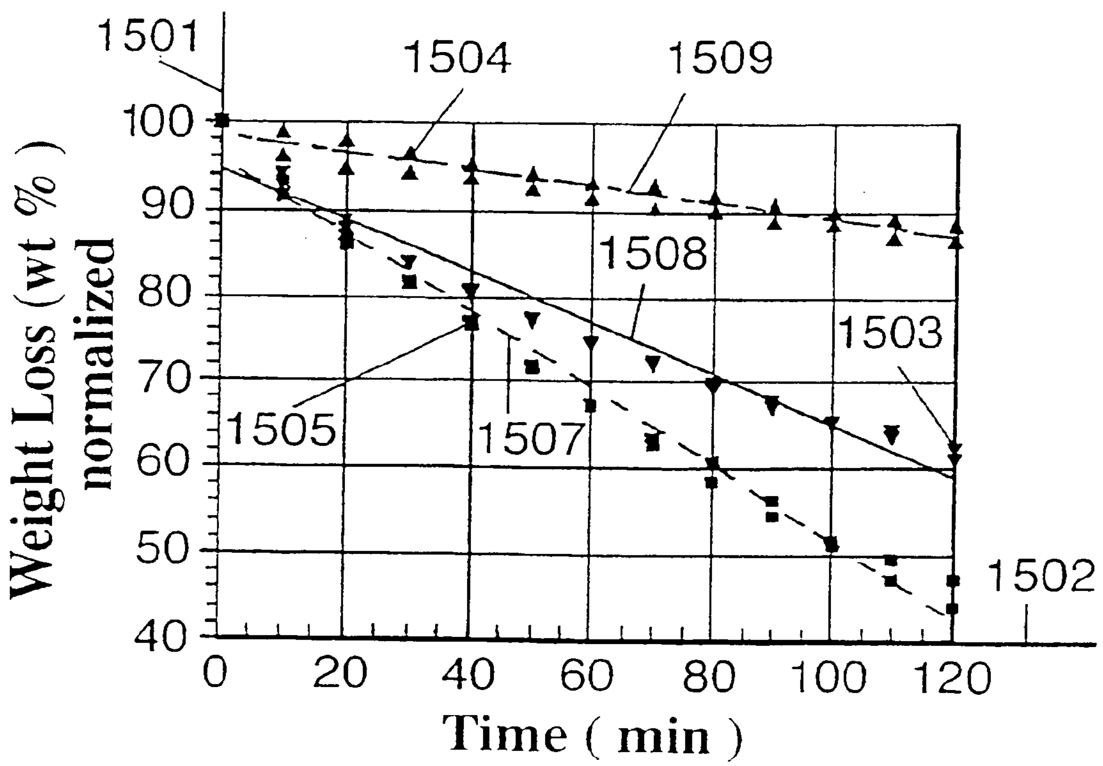

1501
1504
1509
100
90
80
70
60
50
40
Weight Loss (wt %) normalized
1508
1503
1505
1507
1502
0
20
40
60
80
100
120
Time (min)

FIG.16

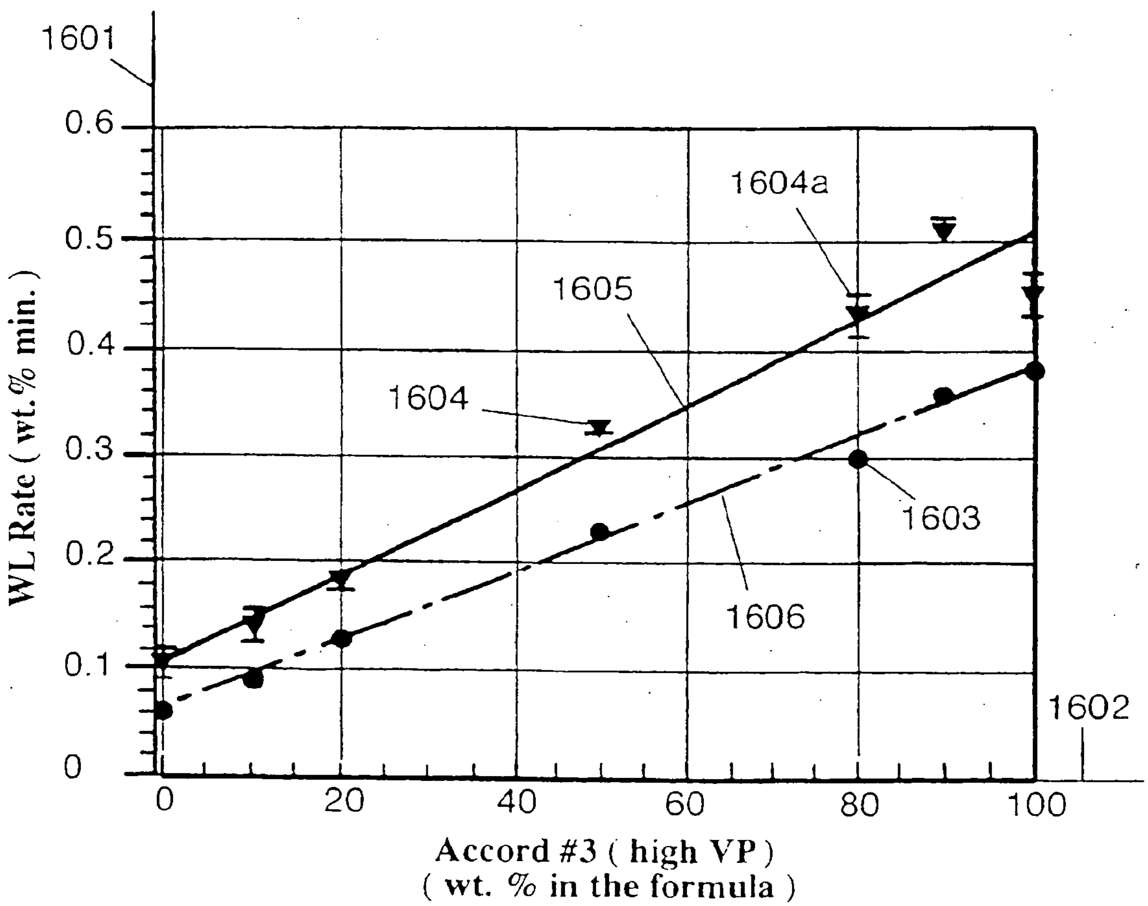

1601
0.6
0.5
0.4
0.3
0.2
0.1
0
WL Rate (wt.% min.)
1604a
1605
1604
1603
1606
1602
0
20
40
60
80
100
Accord #3 (high VP)
(wt. % in the formula)

FIG.17

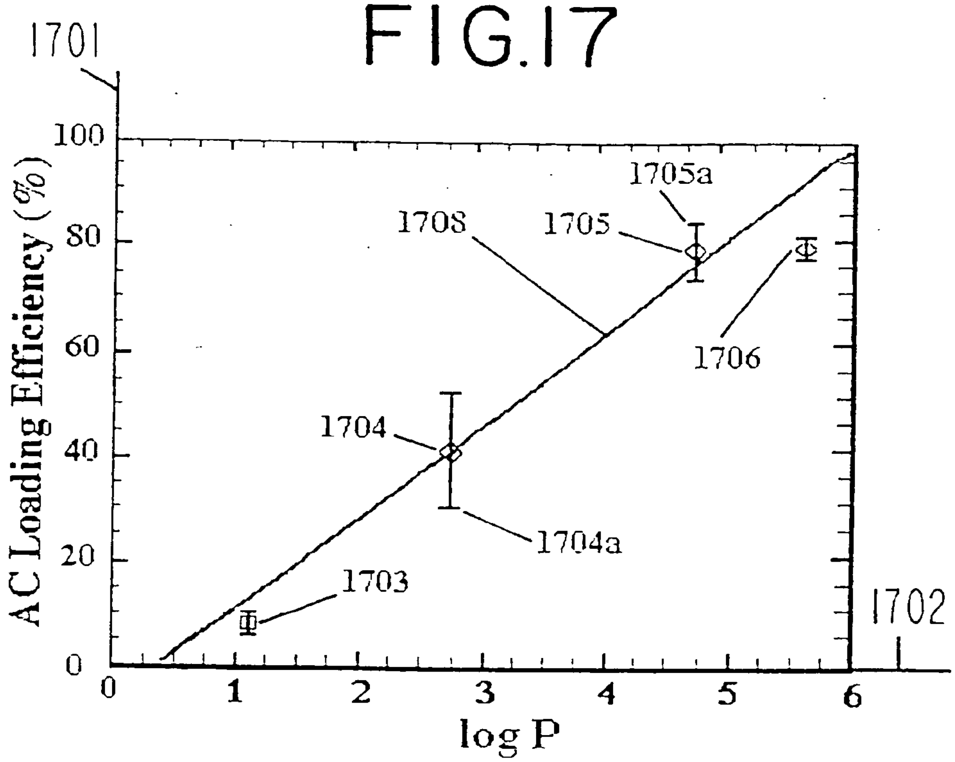

1701
100
80
60
40
20
0
AC Loading Efficiency (%)
1705a
1708
1705
1706
1704
1704a
1703
1702
0
1
2
3
4
5
6
log P

FIG.18-A

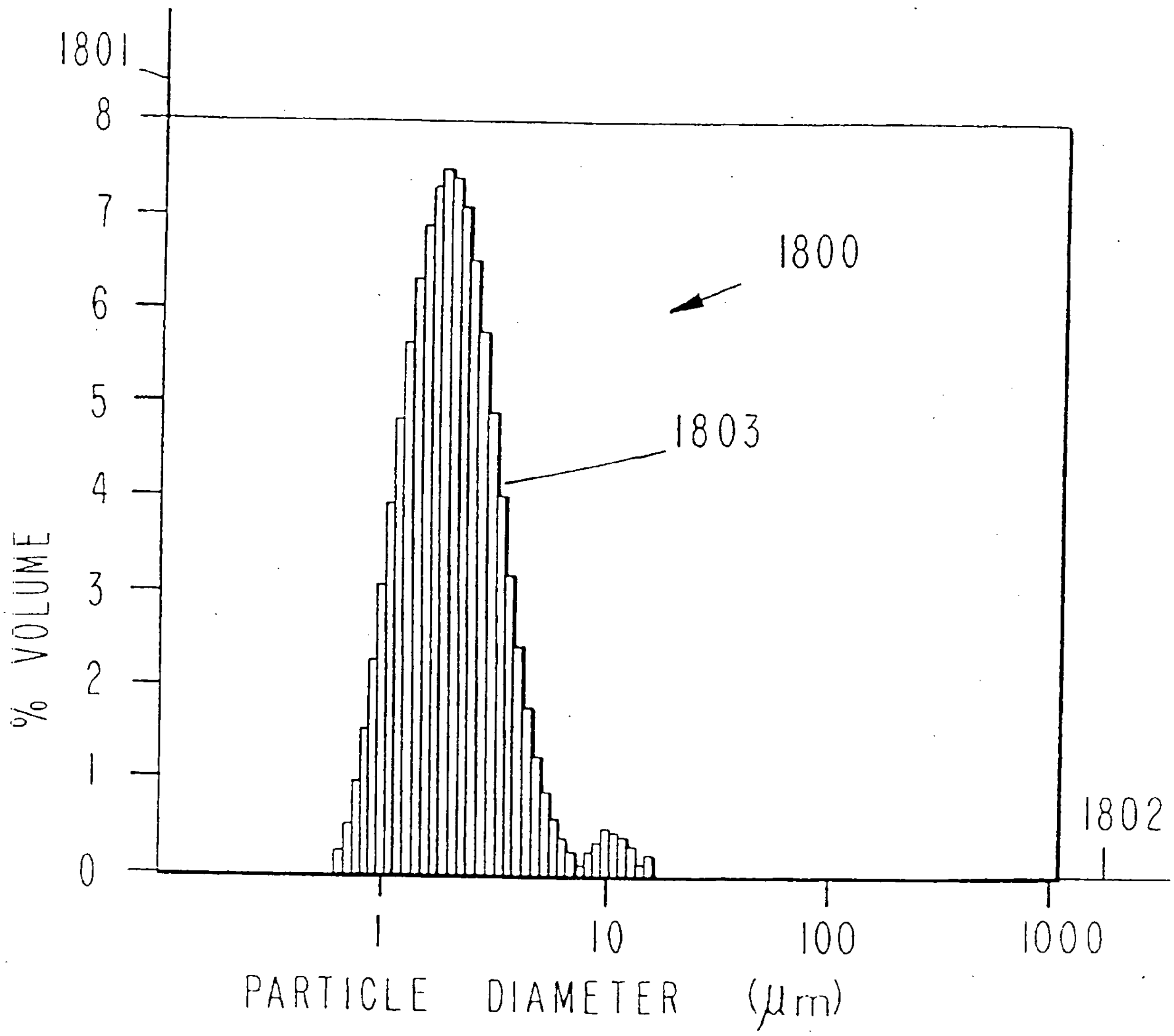

1801
8
7
6
5
4
3
2
1
0
% VOLUME
1800
1803
1802
1
10
100
1000
PARTICLE DIAMETER (μm)

FIG.18-B

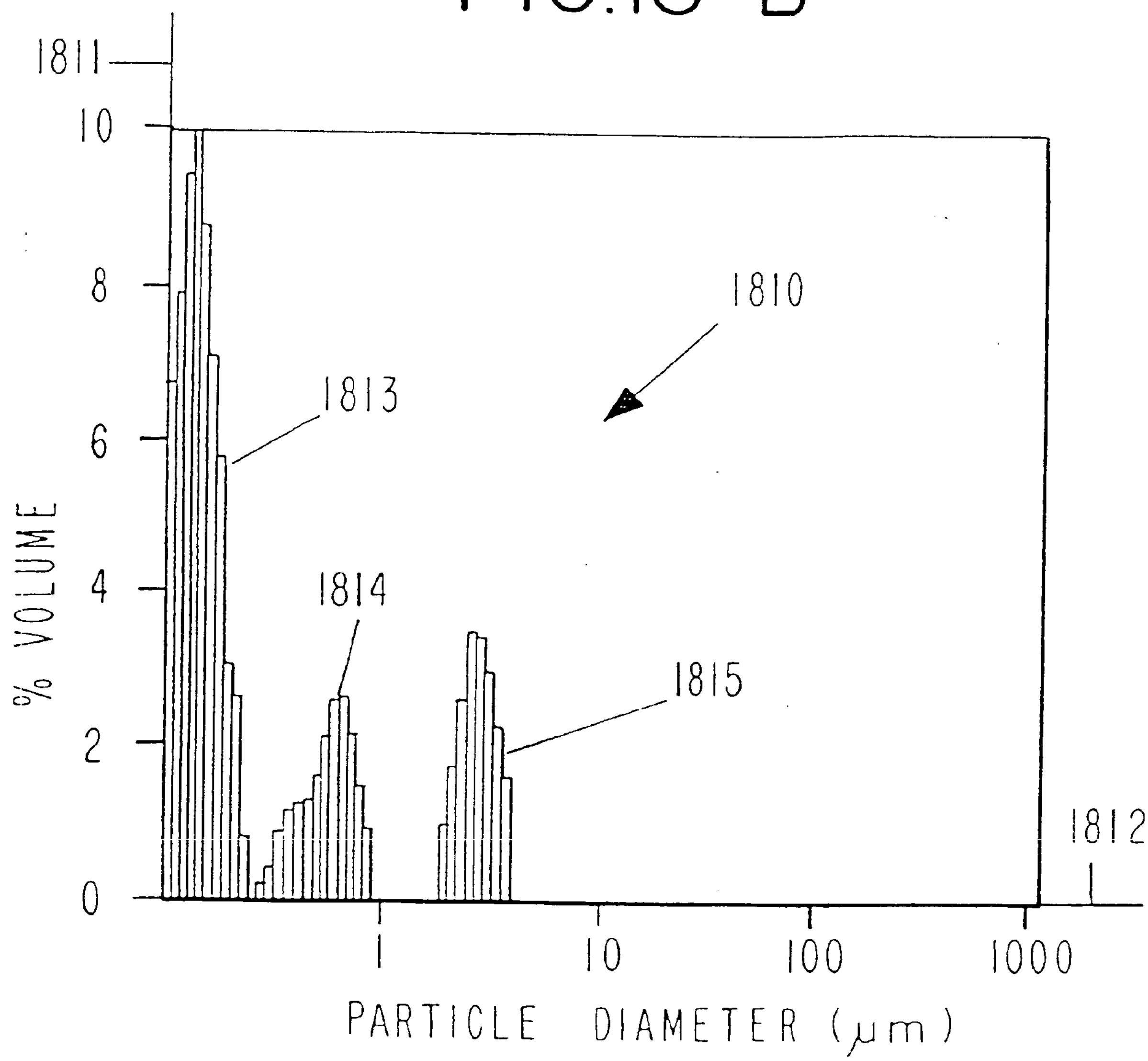
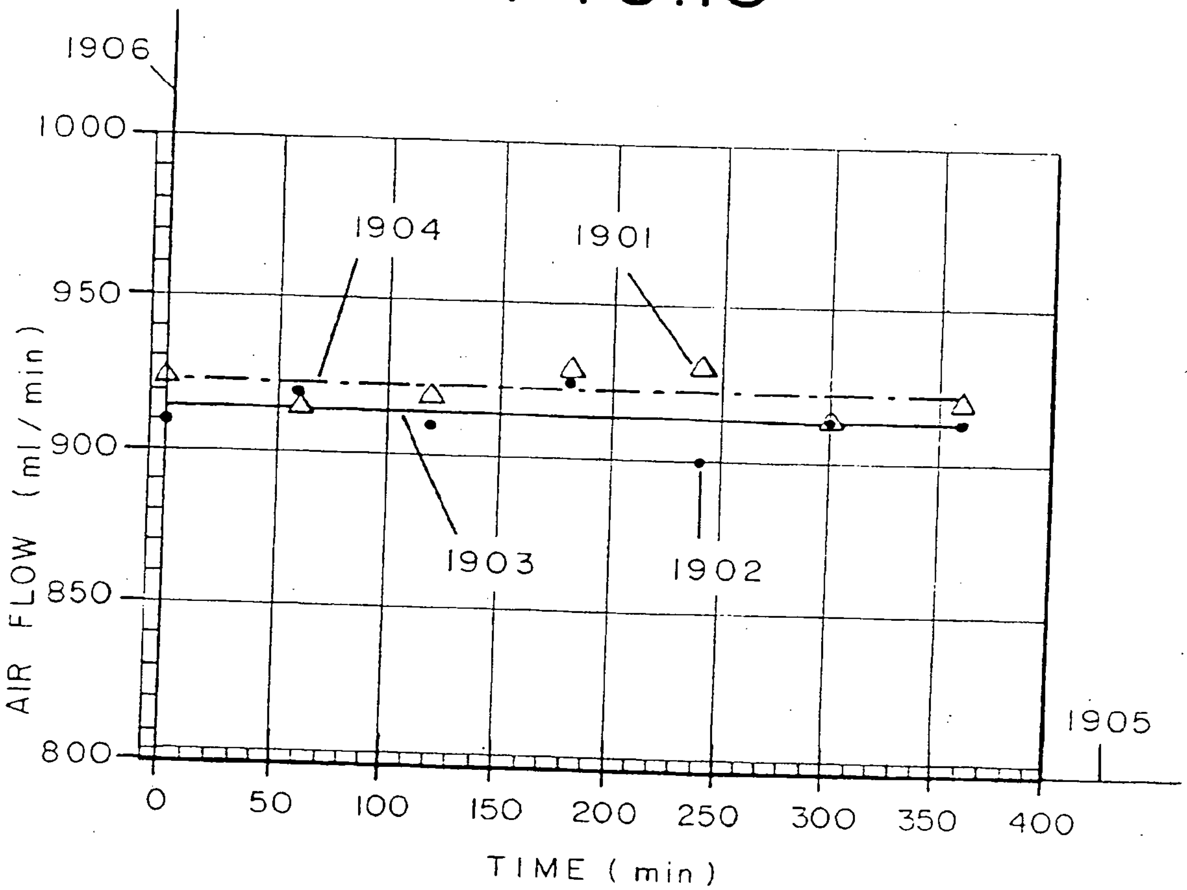

1811
10
8
6
4
2
0
% VOLUME
1810
1813
1814
1815
1812
1
10
100
1000
PARTICLE DIAMETER (μm)

FIG.19


1906
1000
950
900
850
800
AIR FLOW (ml / min)
1904
1901
1903
1902
1905
0
50
100
150
200
250
300
350
400
TIME (min)